Obvious problems in lunar petrogenesis and new perspectives

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Abstract

Forty-six years ago saw the first manned landing on the Moon and the return of the first lunar samples. Since then a vast database has been accumulated with many ideas published on lunar petrogenesis, yet important problems recognized in early days remain under-addressed. In this paper, we first review these problems and emphasize that these problems need resolving before genuine progresses can be made. We then discuss that contrary to the prevalent view, the available data do not show the presence of a strong positive Eu anomaly (Eu/Eu* >1) in the lunar highland crust, but a weak negative one (Eu/Eu* <1) if any. This observation weakens the plagioclase flotation hypothesis, which is the very foundation of the prevailing lunar magma ocean (LMO) hypothesis. Recent success in the determination of abundant water in lunar glasses and minerals confirms the prediction in the early days of lunar research that the Moon may have been a water-rich planet and may still be so in its interior, which disfavours the dry Moon hypothesis, weakens the LMO hypothesis, and questions many related lunar petrogenesis interpretations. Volatilization/degassing (into the vacuum-like lunar "atmosphere") of lunar magmatism during its early histories could have further facilitated plagioclase crystallization and feldspathic crustal formation. The important role and effect of plagioclase crystallization is best manifested by the significant correlation (R^2 = 0.983 for N = 21) of Eu/Eu* (0.24 – 1.10) with Sr/Sr* (0.10 – 1.12) defined by the lunar samples. Although the anorthositic lunar highlands are expected to have large positive Eu (Eu/Eu* > 1; ~ 1.99) and Sr (Sr/Sr* > 1; ~ 2.56) anomalies, yet their absence inferred from the global remote sensing data is best explained by the widespread but areally/volumetrically insignificant KREEP-like material that is enriched in K, rare earth elements and P (hence, KREEP) as well as all other incompatible elements with very strong negative Eu (Eu/Eu* << 1; as low as 0.24) and Sr (Sr/Sr* < 1; as low as 0.10) anomalies. The KREEP-like material may be produced through fractional crystallization enrichment equivalent to processes in advancing, periodically replenished, periodically tapped, continuously fractionated magma chambers. Compared with magmatic rocks on the Earth, lunar rocks are depleted in moderately volatile elements like P, Na, K, Rb, Cs etc., probably associated with volatilization/degassing during the early history of the lunar magmatism. Further work is needed towards an improved understanding of the origin and evolution of the Moon and its magmatism.

"Facts do not cease to exist because they are ignored."
[Aldous Huxley (1894 - 1963)]

1. Introduction

Since the manned landing on the Moon and the first lunar samples returned forty-six years ago, we have learned enormously about the Moon and its petrogenesis as reported in many scientific papers and especially the more recent milestone overviews such as the *New View of the Moon* (*NVOTM*; Jolliff et al., 2006), *Scientific Exploration of the Moon* (Delano et al., 2009), and a rather comprehensive review article with a task specific title *The Moon 35 years after Apollo: What's left to learn?* (Neal, 2009). In this paper, we do not wish to repeat those reviews, but emphasize that some fundamental problems recognized in the early days remain under-addressed because of the prevalent views by the current lunar community with built-in assumptions that have never yet been tested. O'Hara (2000) elaborated in 107 pages on igneous petrogenesis in general and lunar petrogenesis in particular with viable alternatives and convincing arguments, but these have been largely disregarded. We stress that these problems and issues need to be explicitly resolved and alternative interpretations need to be considered towards a genuine understanding of the working of the Moon. Only then are we in the position to proceed on *what's left to learn*.

2. Fundamental problems in lunar petrogenesis and community response

2.1. Long standing problems in the lunar petrogenesis

The senior author (O'Hara 2000, 2001a, 2001b, 2004) has been critical of the 'established' model of lunar petrogenesis. The model includes the following assumptions: the lunar magma ocean hypothesis (LMO), the plagioclase floatation hypothesis, the primary magma hypothesis for mare basalts, and the hypothesis for the presence of a strong positive Eu anomaly in the lunar highland upper crust.

The senior author presents the case that the 'established' standard model is open to challenge on at least six points:

[A]. On the basis of experimental petrology of the lunar samples, the senior author stressed the likely importance of volatilization during lunar lava emplacement (O'Hara et al., 1970; O'Hara, 1972) and suggested that the Moon may have been a volatile (including water) rich planet body at least in its early history, but this likely probability has been disregarded because it is incompatible with the widely accepted "dry Moon" assumption and the lunar magma ocean (LMO) hypothesis by the lunar community(e.g., Wood et al., 1970).

[B]. Io, the innermost satellite of Jupiter, is similar in size and mass to the Moon. Voluminous volcanic eruptions on Io are accompanied by extensive selective volatilisation, but mare-like structures are absent. A comparative study with Io would benefit the research of lunar magmatism and petrogenesis and would make the lunar model more significant and representative in the context of solar system magmatism (O'Hara, 2000).

[C]. The standard lunar model neglects the issue whether the mare rocks represent lava piles or solidified lava lakes. This is important because if the mare rocks are lava piles, they would represent rapidly solidified lunar mantle melts with the composition more directly reflecting the lunar mantle composition, but if they are fossil lava lakes, they would represent slowly cooled and evolved magmas with significant differentiation. In the latter case, caution is necessary when using lunar magma composition to directly infer the lunar mantle composition (O'Hara, 2000).

[D]. The standard lunar model also fails to address the question on the true bulk compositions of the erupted mare basalts. Are they picritic, like a few tens of kilograms of the hand

specimens, or basaltic, like billions of tonnes of the average rock in the mare regoliths (O'Hara, 2000)?

[E]. Alleged primary magmas (basalt hand specimens and picritic glass beads) are interpreted as incipient re-melting products of originally plagioclase-saturated cumulate postulated to have formed from the LMO (Wood et al., 1970; Taylor, 1975). If so, those 'primary magmas' should have plagioclase on their liquidus at the relevant high pressures, but they do not. This implies either the mare basalts are not primary magmas or their source rocks have never seen plagioclase (see below).

[F]. The standard lunar model also avoids discussion of the absence of the requisite strong positive Eu (and Sr) anomaly in the lunar highlands (O'Hara, 2000, 2004), which was the cornerstone of the standard model in its original form and foundation of the *central* LMO hypothesis.

2.2. Absence of lunar community response to these fundamental problems

Multiple working hypotheses, hypothesis testing and scientific debates are logical methodology familiar to and likely welcome by many. In practice, however, this is not the case, and the *bandwagon effect* is prevailing. For example, many researchers are comfortable with following mainstream ideas put forward by some authorities without realizing that the assumptions, hypotheses and interpretations by those authorities are only as valid as those by others without testing and verification. Such widespread tendency has been the real hindrance in science. This seems to be true in all Earth Science subject areas we are familiar with, including the lunar scientific research. The latter is well reflected in the treatment of the few unverified assumptions in the "established" model of the lunar petrogenesis over the past decades as elaborated below.

2.2.1. The hindrance of the unjustified "dry moon" assumption

This concerns [A] above. Volatilization during lunar lava emplacement was proposed over 40 years ago (O'Hara et al., 1970; O'Hara, 1972, 2000), but the "dry moon" assumption accepted as a fact has led to the neglect of the significance of lunar magma volatilization until recently when water in lunar glasses and mineral crystals has been actually measured (Saal et al., 2008; Liu et al., 2012; Hui et al., 2013).

It should be noted that the *NVOTM* (2006) compilation fails to address the issue on selective volatilisation during eruption: (a) 34 years after the proposal and experimental demonstration (O'Hara et al., 1970a,b; O'Hara, 1972), (b) vesicles reported in abundance in mare basalts (e.g., Keszthelyi, 2008), (c) glass beads in the soils identified as fire fountain deposits (pyroclastic) driven by gas of lunar internal origin, (d) 14 years after the glasses demonstrated to display varying S, Na and K abundances as a result of variable volatilization during eruption (Delano and McGuire 1992), (e) 3 years after thermodynamic calculations on the importance of selective volatilisation during eruption (Schaeffer and Fegley, 2003).

The volatiles which drove the fire-fountains included water (Saal et al 2008, 2009, Friedman et al., 2009). If 0.07 wt% water is *still* present in picritic glass of fire-fountain droplets after long flights at high temperature in hard vacuum, how much water was present in the magma before eruption, and how much water in the lunar mantle? Despite this, *NVOTM* (2006) indexes no references to water in the Moon. The effect of water on the melting relationships of lunar materials was explored in the early program (O'Hara et al., 1970a,b; Ford et al., 1972) in order to explain the phase equilibria. Neglecting the presence and effect of volatiles, the "established" lunar petrogenesis model needs reconsideration. Importantly, if the Moon has indeed had abundant

volatiles (i.e., H_2O, F, C, S etc.) in its early histories, or still so in its deep interior, then this requires revision of the "giant impact" hypothesis for the origin of the Moon and the LMO hypothesis. This is because "giant impact" would have removed all (or almost all?) the volatiles from the outset, but abundant volatiles, especially water, remain abundant today in lunar glasses and minerals (Saal et al., 2008; Liu et al., 2012; Hui et al., 2013).

It is also of historical interest to consider the effect on our scientific development as a result of neglecting volatiles. It has been 45 years during which melting experiments should have been undertaken in vacuum, either on terrestrial basalts or on Apollo hand specimens in the large vacuum facility at the Lunar and Planetary Institute (LPI, in Houston) or on a multitude of Shuttle missions, to answer the simple question: Can such melts be exposed to lunar vacuum without fragmenting into droplets under their own vapour pressure? Yet no such study appears to have been done. However, there has been confirmation of rapid alkali loss reported from molten basalt in vacuum (O'Hara et al., 1970a,b; Biggar et al., 1971) by Lewis et al. (1993) who reported experiments at atmospheric pressure (1300-1350 ℃ at Iron-Wustite [IW] fO_2 buffer) on the Knippa basalt. It is an alkali olivine basalt (MgO 12.33 wt.%) with a high liquidus temperature of 1325 ℃, 3.05 wt.% Na_2O and 2.03 wt.% K_2O. At atmospheric pressure the basalt liquid lost ~40% of the initial Na in 4 hours, ~60% in 10 hours, ~75% in 18 hours and ~95% in 72 hours. In vacuum, even faster Na loss of ~85% was observed at 1330 ℃ in 16 hours (Lewis et al., 1993). Most of the initial K was lost in the first 4 hours. The relative rate of Na and K loss is likely variable as a function of melt composition, pressure, temperature and closure of the system.

2.2.2. Missing insights from observations of Io

This concerns [B] above. It is now 36 years since the observations at Io (Smith et al., 1979) that should have alerted the lunar community to the possibilities of selective volatilisation during lunar eruption. However, *NVOTM* (2006) mentioned the possible differences between Io and the Moon, but the obvious relevance of Io to the Moon in terms of eruption environment, and the central issue of volatilization and primary character of the lunar hand specimens, is disregarded.

2.2.3. Missing insights from mare lavas

This concerns [C] above. In order to interpret the geochemical data of basalts as directly indicating the mantle composition, it was proposed by some and accepted by many that most or almost all the terrestrial basalts are rapidly solidified basalts of primary magmas derived from the mantle and erupted on to the surface with no modification (see O'Hara, 2000b). This was considered to be true for the mare basalts by the lunar community. It is probably correct today that most petrologists no longer accept this "primary magma" argument, yet basalt geochemistry is still used as indicating the mantle source composition. The latter approach has been proven to be useful only if the effects of partial melting and magma evolution can be corrected for (e.g., Niu and O'Hara, 2008). Therefore, it is of historical interest, but is also essential to sort out if the mare basalts are of primary nature.

The senior author suggested at the outset (O'Hara, 1970, 1972) and reiterated recently (O'Hara, 2000) that the lunar maria contained within basins are lava lakes (vs. flood basalt piles) which are likely and inevitably to have experienced extensive low pressure gabbro (plagioclase + clinopyroxene) fractionation (see Yoder and Tilley 1962; O'Hara 1965, 1968), resulting in the residual melts represented by the returned mare basaltic samples with a negative Eu anomaly. The "established" flood basalt interpretation automatically treats the mare basalts as primary magmas

with the negative Eu anomaly as reflecting a lunar mantle signature. This discussion of fundamental importance for the lunar petrogenesis has disappeared in the current literature, including *NVOTM* (2006), but the lunar mantle source with a negative Eu anomaly has been widely accepted as a concequence of the LMO hypothesis (see below).

2.2.4. Missing insights from experimental petrology on lunar samples

This concerns [D] and [E] above. The average erupted (vs. hand-specimen) mare basalt compositions were identified experimentally early on (O'Hara et al., 1970, 1972) as being close to low pressure plagioclase-saturated cotectic liquids, indicative of low pressure fractionation and non-primary magma status, resembling terrestrial basalts (including continental flood basalts, ocean island basalts and mid-ocean ridge basalts). However, such low-pressure fractional crystallization demonstrated to take place on the Moon is entirely absent in *NVOTM* (2006).

The 2.28 m mare drill core 70001 to 70009 from the Apollo 17 site (see Fig. 10 of Papike et al., 1998) shows basaltic compositions, but calculations gave 10-30% feldspathic ANT (anorthositic-noritic-troctolitic; highland-derived) gabbros, systematically greater than observed petrographically (see Figs. 1 & 2 of O'Hara 2001b). This points to the likelihood that part of the feldspathic component is occult in the basalt fragments. Further calculations on the *observed* components of sieved coarse fractions of the two most basalt-rich horizons in the core (see Table 1 of O'Hara 2001b) yielded an average basalt composition significantly more feldspathic than the average of hand specimens. All these are consistent with the significance of plagioclase and its crystallization before pyroxene, i.e., close to the low-pressure plagioclase-saturated cotectic liquid compositions as demonstrated experimentally (see Figs 3-6, 9, 10 of O'Hara 2001b).

2.2.5. Missing insights from Eu anomaly in mare basalts

This concerns [E] and [F] above. The lunar magma ocean (LMO) hypothesis is concisely summarized by Day (2006) "*The LMO hypothesis has been built chiefly on the observation that the Moon's anorthositic upper crust has a large positive Eu anomaly, which is complemented by negative Eu anomalies in the mare-basalt source. This relationship is the foundation to notions that the crust may be the result of plagioclase floatation from a large magma chamber (LMO). Numerous models of the differentiation of the Moon involve an initial state in which the outer few hundred kilometers were totally molten. These models can adequately describe the anorthositic crust and the constant enriched incompatible-element signature of KREEP, the residual liquid from the LMO differentiation event.*"

This means that if the alleged Eu anomaly does not exist or its origin is incorrectly explained, the plagioclase flotation model for the lunar anorthositic crust needs revision and the LMO model needs reconsidering accordingly. Indeed, the straightforward explanation for the negative Eu anomaly (i.e., Eu/Eu* <1) in the mare basalts is plagioclase fractionation as manifested by plagioclase microphenocrysts in lunar hand specimens (O'Hara et al., 1974) and experimental demonstrations (see above). However, much of the lunar community since 1971 has excluded low pressure gabbroic fractionation as the cause of the negative Eu anomalies in the mare basalts, but insisted on the negative Eu anomalies inherited from the mare mantle source which is the cumulate from the LMO and whose negative Eu anomalies are complemented by plagioclase floating in the LMO to form the anorthositic upper crust (e.g., Grove, and Krawczynski, 2009; also in *NVOTM*, 2006).

3. The prevailing lunar magma ocean (LMO) hypothesis and its validity

The LMO hypothesis has been widely perceived as the central theory for lunar petrogenesis. As stated above, the motivation for the LMO hypothesis was simply the need for an environment in which volumetrically significant plagioclase can be crystallized and float in such an LMO to from the lunar highland crust with a positive Eu anomaly (i.e., Eu/Eu* >1). Meanwhile, the LMO would precipitate the mafic cumulate whose partial melting imparts the negative Eu anomaly (i.e., Eu/Eu* < 1) in the mare basalts. The later interpretations has been admitted by *NVOTM* (2006) to be the sole explanation for the negative Eu anomaly in the mare basalts.

The LMO hypothesis has two testable predictions: (1) there should be a prominent positive Eu (and Sr; see below) anomaly [F] in the average lunar crust composition by 3.9 Ga, when mare basalts were sampling the lunar mantle. However, this is shown not to be the case (O'Hara 2000); (2) low mass fraction re-melting products of cumulates, which were precipitated at high pressure from plagioclase-saturated magmas, *must* have plagioclase as an early crystallising phase at low pressure – yet the alleged primary magmas from the LMO never have plagioclase as an early crystallising phase at any pressure (see Grove and Krawczynski, 2008). These observations and reasoning question the validity of the LMO hypothesis (see below).

4. Latest statements of the Lunar Community

A comprehensive review by Neal (2009) and a collection of state-of-the-art views on lunar petrogenesis in an issue of *Elements "Scientific Exploration of the Moon"* led by Delano (2009) continue to advocate the LMO idea as well as supportive assumptions, while neglecting issues and alterative interpretations in the literature as reviewed above. Also, in places of allocating priority for some observations (i.e., the senior author's original work), references are rather selective, perhaps because the alternative interpretations differ from the standard models although this is not a scientifically acceptable practice. In any case, we welcome balanced and objective discussion towards a genuine understanding of the working of the Moon.

4.1. The LMO hypothesis remains a to-be-tested hypothesis

Delano (2009) states "*The ferroan anorthosites are likely to have been formed during the global magma-ocean event associated with the Moon's rapid accretion. Plagioclase flotation in that magma ocean is widely considered to have been part of the process that led to the formation of the ferroan anorthosites at ~4450 Ma. The magnesian and alkali suites were formed later, but prior to ~3800 Ma, by later melting of cumulates formed during the magma-ocean event.*" While this summary statement as stated by Day (2006; see above) is useful, it treats LMO hypothesis as fact, which is not, but remains an assumption and a hypothesis to be tested. Taylor (2009) reiterates the prevailing view that the negative Eu anomaly in the mare basalts resulted from melting of the lunar mantle of cumulate origin produced by magma ocean and plagioclase floatation. He emphasizes "*… While we do not understand all the processes that could have operated in such a huge magma system, it seems clear that the magma ocean set the stage for all subsequent magmatic activity on the Moon*". Grove and Krawczynski (2009) show the LMO in the firm of cartoons as the most effective model to explain the lunar petrogenesis.

We should emphasize that the "dry moon" assumption has failed because of the confirmation of a volatile-bearing (if not rich) Moon (e.g., O'Hara et al., 1970; O'Hara, 1972, 2000) by recent studies (e.g., Saal et al., 2008; Liu et al., 2013; Hui et al., 2013). It follows that the giant impact hypothesis is in doubt as there would be otherwise no (or almost) volatiles left. It follows still that

if the giant impact hypothesis needs reconsideration, then the LMO hypothesis also needs reconsideration because of inadequate energy for global melting.

4.2. The primary magma interpretation for mare basalts remains an assumption-based speculation

Delano (2009) states mare basalts to be of primary magma and of flood basalt origin without explaining that these are only favoured assumptions. Grove and Krawczynski (2009) identify the situation highlighted in [E] above that overlooks the related problem. We emphasize that the first drop of melt produced on reheating of an assemblage of minerals which originally precipitated from a plagioclase-saturated liquid *must* itself be plagioclase-saturated, regardless whether the plagioclase crystals originally present are still there or not (see below). This assertion needs qualifying considerations, hence we use the wording in [E] "Those 'primary magmas' should have plagioclase *close to their liquidus at the relevant high pressures* – they do not." This phase equilibrium reasoning requires serious testing of the hypothesis whether the source rocks of the mare basalts are indeed LMO cumulate after plagioclase removal.

4.3. Evidence for non-primary lunar mare basalts vs. the standard primary magma model

Delano (2009) and Grove and Krawczynski (2009) give credit only to Ringwood and Essene (1970) for the low pressure phase equilibria in Apollo 11 basalts. In fact, the latter authors reported only results for a single synthetic composition matching the hand specimen average composition run in iron capsules at unknown oxygen fugacities. On the other hand, O'Hara et al. (1970a) reported 6 results at the controlled oxygen fugacity of the IW buffer for natural sample 10017 (hand specimen) and 5 results for sample 10084 (regolith). O'Hara et al. (1970a,b) correctly established an ulvöspinel liquidus above 1202 °C, olivine and ilmenite entry between 1188 and 1162 °C, plagioclase entry between 1133 and 1099 °C and solidus close to 1070 °C in 10017 and a plagioclase liquidus above 1188 °C and solidus close to 1100 °C in 10084 (with liquidus-solidus intervals of >130 °C and ~100 °C, respectively).

All the above, in addition to results for 13 synthetic analogue samples at two controlled oxygen fugacities, established that the Apollo 11 basaltic compositions lay close to those of low pressure cotectic liquids saturated simultaneously with ulvöspinel, ilmenite, pseudobrookite (armalcolite), olivine, clinopyroxene, pigeonite and plagioclase at a temperature of ~1115 °C ± 15 °C. These observations demonstrated convincingly that Apollo 11 samples do not represent primary magmas, but cotectic compositions resulting from extensive low pressure fractional crystallisation (O'Hara et al., 1970a,b). Walker et al. (1974) confirmed the experimental work of O'Hara et al. (1970a,b) for a near-cotectic relationship for these compositions, yet Ringwood and Essene (1970) preferred the cotectic compositions to be of low-pressure primary magmas derived from mantle compositions of olivine pyroxenite without fractional/equilibrium crystallization.

We should note the potential experimental problems in studying the lunar samples. The O'Hara laboratory in Edinburgh was the *only* principal investigator group appointed to carry out experiments on Apollo 11 natural samples prior to that mission, although several other workers both within and outside the Apollo scientific program made enthusiastic competitive attempts to explore the phase equilibria, each in their own synthetic analogue of the average Apollo 11 basalt hand specimen composition published by the Lunar Sample Preliminary Examination Team (LSPET, 1969), yet most of them were unaware of the experimental difficulties. O'Hara (2000) gave a thorough review of phase equilibria concerning the above, relevant to lunar high TiO_2 basalt, fO_2 values achieved in experiments in iron capsules such as those reported by Ringwood

and Essene (1970), their effects on charge composition and observed phase equilibria (also see Figs. 4, 5 of Ford et al., 1972) including multiple saturation, as well as providing a thorough evaluation of the minor extent of side-effects in the molybdenum capsule technique when properly employed.

4.4. Selective citations of preferred interpretations and additional evidence for non-primary lunar mare basalts

Grove and Krawczynski (2009) allocate credit for determination of phase equilibria in mare basalts at higher pressures exclusively to one laboratory, stating "*Experimental studies of the melting relations at high pressure and high temperature of the Apollo 11 (Ringwood and Essene 1970) and Apollo 12 (Green et al. 1971) lavas showed that Mg-Fe silicates were the first minerals to crystallize when the mare basalt cooled to its liquidus. At low pressures, olivine was the first liquidus phase, and at higher pressures, pyroxene was the first phase to crystallize;*" As noted above, Ringwood and Essene (1970) did not use natural samples, while O'Hara et al. (1970a,b; Table 1 and Fig. 1) provided data for 28 runs on mare basalt 10017 and 33 runs on regolith 10084 at pressures ranging from 0.2 to 3.7 GPa which revealed a more complicated situation than conveyed in the quoted text, yet this comprehensive and careful earlier work by the senior author is ignored. High TiO_2 basalt *hand specimen composition 10017.51* had a TiO_2-rich oxide phase on the liquidus (O'Hara et al., 1970a) at low pressure, joined by olivine then by clinopyroxene and finally by plagioclase as olivine is resorbed, forming an ilmenite-gabbro assemblage. Increasing pressure toward that of ~200 km depth in the Moon causes clinopyroxene to crystallize at higher temperatures with the olivine field suppressed. At depths greater than ~200 km, clinopyroxene is the first liquidus phase. As a candidate for primary magma status, sample 10017.51 would require extraction from an ilmenite-clinopyroxenite mantle source at 200 km, 1230 ℃ or titaniferous clinopyroxenite at greater depths, but never an olivine-pyroxenite. Plagioclase is never encountered close to the liquidus in this composition and the phase diagram illustrates the expansion of the clinopyroxene liquidus field at the expense of the plagioclase and Ti-oxide fields with increasing pressure. These relationships demonstrate that liquids produced by small mass fractions of partial melting of originally plagioclase, olivine and ilmenite-saturated cumulates are richer than 10017.51 in potential plagioclase, ilmenite (at depths greater than 200 km) and olivine and poorer in clinopyroxene.

Grove and Krawczynski (2009) continue, "… *Plagioclase should be on the liquidus if it was present in the source region during the melting event, as the Eu anomaly seemed to indicate, yet olivine and pyroxene were the only high-pressure liquidus phases. This evidence strengthened the arguments that the Eu-rich lunar crust was involved in a Moon-wide melting and differentiation event. Thus, the source of the mare lavas was composed of minerals that had been in equilibrium with plagioclase when they crystallized and had inherited their negative Eu anomaly from this prior magma-ocean-processing event*." Obviously, this long-held preconception on the LMO assumes mare basalts to be primary magmas while neglecting evidence for their non-primary magma nature as expected and as demonstrated experimentally (O'Hara et al., 1970a,b; O'Hara, 1972, 2000; Walker et al. (1974) .

We should also clarify further the significance of multiple saturation. O'Hara (2000; see p. 1555 and notes 99-102, p 1606) identified a problem about lunar picritic glasses. If these glasses represented primary magmas, they would define a hot, supra-adiabatic thermal gradient persisting until late in mare volcanic history in a putative cumulate mantle which would have been gravitationally unstable throughout. More generally, O'Hara and Herzberg (2002) observed

"*There is nothing special about the simultaneous appearance of olivine and orthopyroxene on the liquidus of a primitive basaltic composition. It is a necessary, but not sufficient, condition to support the argument that a particular composition is a primary magma produced by equilibrium (batch) melting of a harzburgite residue at a particular pressure and temperature.*" Boninite liquids with both olivine and orthopyroxne as the common liquidus phases are best terrestrial examples of partial melting a harzburgite source (Niu, 2005). Another problem is the control of composition, for which Grove and Krawczynski (2009) indicate the significance of oxygen fugacity, which is likely to be true, but there are likely greater variations in source compositions to be considered (Friedman et al., 2009).

4.5. Summary

The *LMO hypothesis* is the very centre of all aspects of the lunar petrogenesis. The giant impact may have led to the whole-moon scale melting and the LMO formation. Flotation of plagioclase crystallized from the LMO formed the anorthositic lunar highlands. The LMO also precipitate dense mafic cumulate to form the lunar mantle, whose later re-melting produced mare basalts. Because Eu, relative to other REEs, is compatible in plagioclase, the anorthositic lunar highlands thus have a significant positive Eu anomaly. As a result, the lunar mantle of cumulate origin from the LMO is expected to have a negative Eu anomaly, which as the source of the lunar mare basalts impart the negative Eu anomaly in the mare basalts. While this interpretation seems reasonable, it has a hidden assumption that all the mare basalts are *primary magmas* without undergoing any modification once produced. The validity of the *LMO hypothesis* needs testing and the primary lunar mare magma assumption needs verifying also. While anorthositic rocks are expected to have positive Eu anomaly, whether he lunar anorthositic highland upper crust has a strong positive Eu needs examination. If one of the above is proven to be questionable, the whole "established" lunar petrogenesis model needs reconsideration. Furthermore, if the "dry moon" assumption fails, the impact hypothesis for the origin of the moon fails also. As a result, the LMO fails accordingly. There is no doubt that magmas can preserve some source characteristics (Niu and O'Hara, 2008), but this is not the proof that the erupted mare basalts are primary magmas without undergoing cooling and modification during scent. We have noted for long that primary magmas do not exist in nature and can only be found in experimental charges (Niu, 2005). In fact, cooling induced magma evolution and magma chamber processes, especially at low pressure, dominate the compositional characteristics of basaltic lavas on Earth, also of the mare basalts (O'Hara, 1977; Walker et al., 1979; O'Hara, 2000; O'Hara and Herzberg, 2002; Niu and O'Hara, 2008), as discussed above and furthermore below.

5. Lunar surface compositions: returned samples, meteorites and remote sensing compositions and the insights they offer

Returned lunar samples are direct materials for studying the petrogenesis of lunar rocks and the composition and evolution of the Moon, but such materials are rare and whether they represent compositions of the morphological features sampled from or very localized phenomena remain unknown. Hence, their significance is subject to interpretation and speculation. Therefore, indirect means such as remote sensing to measure lunar surface chemical compositions of global coverage become essential and more objective in correlating chemical compositions with lunar surface morphological features despite some uncertainties (see assumptions below).

Remote sensing of the lunar surface and upper crust has been provided by photogrammetry, laser altimetry, radar sounding, seismic studies, gravity measurements, reflectance studies in the

infra-red to ultra-violet, X-ray, Gamma-ray and neutron spectrometry. Data have been obtained from earth-based telescopes, orbiting Apollo command modules, Mariner 10, Galileo, Clementine and Lunar Prospector. The geochemical data measure the local surface regolith composition and are calibrated against returned regolith samples from nine sampling sites (ground truthing). *NVOTM* (2006) provides an in-depth consideration of the many factors involved in the assessment of remote sensing data on the composition of the lunar surface and subsurface.

5.1. Assumptions relevant to interpretation of remote sensing data

Assumption 1 [A1]: The earlier explicit assumption back in 1969 was that the hand specimens from Apollo 11 represent samples of solidified primary magmas. [A1] was later extended to the more mafic hand specimens from other mare landing sites.

Assumption 2 [A2]: The regolith developed over the maria *is not* representative of the immediately underlying lava flows in order to support [A1] because the regoliths show systematic differences (documented since 1969) from "primary magmas" represented by mare hand specimens.

Assumption 3 [3A]: The regolith developed over all highland regions *is truly* representative of the immediately underlying bedrock. This was required for the positive Eu anomaly that should exist in the anorthositic highland crust. Hand specimens from the highlands do have a positive Eu anomaly because of the anorthositic composition (e.g., Lucey et al., 2006), but the average lunar highland regolith does not, and actually has a small negative Eu anomaly (O'Hara, 2000; also see below).

Assumption 4 [A4]: This is revised from [A3] that the regolith developed over all highland regions *is not representative* of the underlying lunar crust - which permits the further model-saving assumption 5 [A5] that the requisite positive Eu anomaly must still be present, but buried beyond detection at some depth within the crust.

[A4] is consistent with [A2] in that hand specimens, whether collected from the maria or highlands, do not represent the mean compositions of the surface or the underlying strata. [A2] is valid only if the relatively feldspathic nature of the mare regoliths is due to vertical mixing of underlying highland crustal material into the surface regoliths.

With all the observations considered a revised assumption is needed, [A3*]: Regolith compositions everywhere are dominated by immediately underlying bedrock, with both lateral and vertical mixing possible, but limited (Cahill et al., 2009).

5.2. Lunar surface composition and the LMO hypothesis

Magma oceans could have once existed on the Earth, Mars and the Moon if the Moon were indeed formed as a result of Mars-size planet impact with the proto-Earth because of enormous energies involved (e.g., Brandon, 2007). Because the LMO remains a hypothesis to be tested, it is logical to examine the cornerstone assumptions for the LMO hypothesis. As summarized by Day (2006; see above), the proposal for the LMO was the need to explain the compositional differences of returned lunar materials between the anorthositic lunar highlands and the basaltic lunar Maria (e.g., Wood et al., 1970). If a global LMO existed in the very early history of the Moon, the buoyant plagioclase crystals would float to form the anorthositic highland crust with a strong Eu anomaly. The dense mafic minerals would sink to form the cumulate mantle with a negative Eu anomaly. Re-melting of this cumulate mantle later would produce the mare basalts with inherited negative Eu anomaly. The incompatible element highly enriched KREEP (abbreviation of potassium [K], rare earth elements [REE] and phosphorous [P] enriched materials) samples would be interpreted

as resulting from protracted fractional crystallization. These are illustrated in the "whole-Moon" normalized REE pattern diagram (Fig. 1). Obviously, the LMO hypothesis offered a convenient explanation for the lunar petrology and geochemistry on returned lunar samples, but what could have caused the formation of the LMO? A giant planetary impact was assumed to have created the LMO (see Wood, 1972; Halliday, 2000; Brandon, 2007). Hence, what we know about the petrogenesis of Moon rocks is a chain of assumptions with the objective of interpreting the observations, starting from lunar surface compositions. The question is whether the observations are all valid and whether the interpretations are unique. We can examine the available lunar data on returned samples and global lunar remote sensing data.

5.2.1. Basic observations

Figure 2 compares, in terms of REEs, returned samples from lunar highlands and Maria using the highland ferroan anorthosite (FAN) and KREEP as extreme references. It is apparent that the Eu/Eu* increases with decreasing overall abundances of non-Eu REEs.

Figure 3 shows that the comprehensively representative 21 lunar surface samples (soils) from 9 Apollo and Luna sites (from the compilation by Lucey et al., 2006) altogether manifest:

(1) the rather similar flat incompatible trace element patterns;
(2) the varying abundance levels;
(3) the characteristic depletion (or negative anomalies) of Rb, K, Sr, P and Eu (also Cs not shown) compared with Earth rocks;
(4) lacking significant fractionation of more incompatible elements (towards left) from less incompatible elements (towards right).

If these lunar surface samples are indeed magmatic products, then the lunar magmatism differs significantly from the Earth magmatism. The following are possible explanations for the data in Figure 3:

(1) The characteristic depletion of Sr and Eu could be due to plagioclase fractionation (see Niu and O'Hara, 2009; also see below);
(2) The Lunar mantle may be characteristically depleted in Rb, K, P and Cs relative to their adjacent incompatible elements;
(3) The elevated abundance levels (vertical variation) for a given element is apparently consistent with varying extent of fractional crystallization (e.g., Niu and O'Hara, 2009);
(4) Compared to Earth rocks, the rather insignificant inter-elemental variation of incompatible element abundances as a function of increasing incompatibility (from right to left) may suggest a relatively uniform lunar mantle source composition with little variation in relative extent of melting;
(5) The varying incompatible element abundances may also suggest "magma mixing" between highly evolved KREEP-like melts enriched in incompatible elements with anorthosite-like cumulate depleted in incompatible elements;
(6) The mixing mentioned in (5) could be "mechanical" mixing (see below).

Figure 4 confirms the above notion that the lunar magmatism differs from the Earth magmatism. The significant positive linear correlations of REEs (e.g., Ce) with all other incompatible elements mean that they are similarly incompatible during their magmatic petrogenesis. Source heterogeneity and varying extent of melting can be ruled out as the cause of the observed lunar surface compositions in Figure 3 because of the largely sub-parallel patterns if

we assume the elemental behavior (e.g., the relative incompatibility of elements) during lunar magmatism is the same as or similar to the Earth magmatism if the source mineralogy is similar. Varying degree of fractional crystallization, which can explain the elevated incompatible element abundance levels, can also be ruled out as the cause because this abundance variation is uncorrelated with major elements (see below). It is important to note that compared to Earth rocks, K, P, Rb and Cs in lunar samples are highly depleted or have "negative" anomalies (Fig. 3), yet these elements show similar incompatibility to all other incompatible elements as manifested by the similarly significant linear correlations (Fig. 4). That is, the depleted signature of these elements could be interpreted as inherited from the lunar mantle source that differs significantly from the Earth's mantle. This conclusion is of far-reaching significance. However, caution is necessary for such a conclusion before we actually understand the significance of Figures 3 and 4. As will be clear later, the best explanation for the data is more like a "mechanical" "magma mixing" between a highly enriched KREEP-like melt with highly depleted anorthosite or depleted mafic cumulate (see below).

5.2.2. The significance of Eu anomalies

Because the presence of a substantial positive Eu anomaly in the average pre-3.9 Ga lunar highland crust is the cornerstone of the 'established' lunar petrogenesis model (see above), it is important to examine the issue of the Eu anomaly.

On the basis of the returned samples (both highland anorthositic and mare basaltic rocks) from a number of sites, Korotev and Haskin (1988) demonstrated that the Eu anomaly can be well estimated by using Sm, Eu and Th concentrations and by correlating Sm/Eu ratios with Th. They then obtained a mean Th value of 0.91ppm for the highlands surface crust acquired by the orbiting gamma-ray experiments. The mean concentration of Sm in the lunar surface crust was found to be between 2 and 3 ppm, and that of Eu between 0.7 and 1.2 ppm. The results indicate that there is no significant enrichment or depletion of Eu, compared to Sm, relative to chondritic abundances; i.e., there is no significant "Eu anomaly" in average highland upper crust, contrary to predictions by some earlier studies (see O'Hara, 2000, 2004 for review).

We apply a more robust method to evaluate Eu/Eu* values of lunar samples shown in Figure 5, where the representative soil samples from all Apollo and Luna sites (red dots) and lunar highland samples reported in the Lunar Source book (thick blue line). Both show statistically significant power-law relation for Th content and Eu/Eu*. Clearly, Th <~0.7 ppm when Eu/Eu* >1, whereas Th >~0.7 ppm when Eu/Eu* <1. Because all these samples indeed represent the lunar surface material, we can objectively state that the lunar surface area (if not volume), especially the lunar highlands, with a positive Eu anomaly is rather limited. In fact, O'Hara (2000b, 2004) noted the association of a positive Eu anomaly with lunar highland anorthosite samples, but demonstrated no such a positive Eu anomaly present for the lunar highland crust as a whole. If anything, there is a negative anomaly.

Figure 6 shows the frequency of Th concentrations from gamma ray spectrometry (GRS) for the whole lunar surface. Conspicuously, on a global scale the lunar surface with Th <0.7–0.8 ppm is rather small, probably <~1%. That is, the lunar surface area (rocks and regoliths) with a positive Eu anomaly is areally <1% (Fig. 5). Without doubt, the plagioclase-rich rocks like the varying types of anorthosite must have a positive Eu anomaly (Fig. 2; also see Niu and O'Hara, 2009). However, the data (Figs. 5, 6) indicate that the lunar highlands, although areally expansive and claimed to be dominated by anorthositic rocks, must have on average a negative, not *positive* Eu

anomaly. Hence, a positive Eu anomaly based hypotheses on the lunar highlands are questionable, including the plagioclase flotation hypothesis, and hence the prevailing LMO hypothesis.

In the context of this discussion, Korotev (2005) dismissed the argument of O'Hara (2000, 2001b) as 'misconstrued', but the argument is straightforward. If the lunar mantle sources of Apollo 11, 12, 15 and 17 basalts had a negative Eu anomaly at 3.9–3.5 Ga and there is no positive Eu anomaly in the average pre-mare crust, where is the compensating reservoir with a positive Eu anomaly? If truly anorthositic portion of the lunar highland crust is all that remains of the 'original' plagioclase flotation crust, whence comes the vast mass of highly feldspathic pre-3.9 Ga highland materials with negative Eu anomalies? Because plagioclase is the least dense silicate mineral and its floatation is the central tenet of the LMO hypothesis (Wood et al., 1970), then the floating anorthosite must all be preserved on the surface. The farside lunar highlands have been considered to consist of the floating anorthosite (e.g., Taylor, 2009), yet this is inconsistent with the data in Figures 5 and 6 if we accept the assumption that plagioclase-rich rocks necessarily have a positive Eu anomaly. The returned Ferroan Anorthosite Suite (FAN) samples indeed have low Th <0.15 ppm (Taylor, 2009) which favours the presence of a positive Eu anomaly, but the far majority of the LP GRS data have Th >0.7 ppm (Figs 5 and 6; also see Taylor, 2009). Hence, from REE and Th data on returned samples and also global LP GRS data, we cannot avoid the conclusion that the lunar highland crust has no positive Eu anomaly, which is highly paradoxical against the plagioclase flotation and the LMO hypotheses.

The obvious paradox can be resolved by answering either or both of the following questions: (1) Is the anorthositic crust is areally (or volumetrically?) rather small on the whole-Moon scale? (2) Does the anorthositic crust contain widespread KREEP-like material with a strong negative Eu anomaly? For the latter, is it possible that the KREEP-like material on the lunar highlands may be underestimated because of sampling bias yet objectively recognized through the LP GRS remote sensing method?

5.2.2. Insights from Sr and major element data–two and three end-member mixing

If the Eu anomalies, whether positive or negative, in the lunar rocks are indeed caused by processes involving plagioclase accumulation or fractionation, then these anomalies should also be conspicuous for Sr too, because plagioclase is the single rock-forming silicate mineral that can effectively and simultaneously fractionate both Eu and Sr from otherwise similarly incompatible elements (Niu and O'Hara, 2009), i.e., Eu from Sm and Gd ($Eu/Eu^* = Eu_N/[Sm_N*Gd_N]^{0.5}$) and Sr from Pr and Nd($Sr/Sr^* = Sr_N/[Pr_N*Nd_N]^{0.5}$). Indeed, Niu and O'Hara (2009) demonstrate that Eu/Eu* and Sr/Sr* define a curvilinear trend in MORB melts as a straightforward consequence of plagioclase fractionation (Fig. 7) because both Eu/Eu* and Sr/Sr* in the evolving MORB melts also linearly and systematically decrease with decreasing MgO (see Fig. 4 of Niu and O'Hara, 2009). Figure 7 shows indeed that the lunar samples (Lucey et al., 2006; Saal et al., 2008) do define a significant linear trend that is most consistent with the effect of plagioclase by its addition (increasing Eu/Eu* and Sr/Sr* ratios) or subtraction (decreasing these two ratios). However, the linear trend differs from the plagioclase fractionation trend defined by MORB melts (Fig. 7), but is most consistent with a mixing trend.

From Figure 2, we can predict that if we consider Eu anomalies and REEs, one end-member may be KREEP-like material highly enriched in incompatible elements (Fig. 3) including Th (Fig. 5) with a strong negative Eu anomaly (hence also negative Sr anomaly with $Sr/Sr^* << 1$). The anorthositic cumulate rocks depleted in incompatible elements (Fig. 3) including Th with a strong positive Eu anomaly (hence positive Sr anomaly with $Sr/Sr^* >> 1$) would be the other end-

member. We should note that the constructed "upper few kilometres (FUpCr) of typical feldspathic crust" based on feldspathic lunar meteorites" by Korotev et al. (2003) has very high Eu/Eu* (2.02) and Sr/Sr* (2.54), located remotely on the same trend linearly projected from the data. Hence, this FUpCr would be the ideal feldspathic end-member (see Lucey et al., 2006). All this analysis actually confirms that mixing between these two end-members explains all the lunar *incompatible trace data* presented in Figures 2-5 and 7.We should also note that the glass beads (melts) plot on the same mixing trend in Figure 7. The mixing could be melt-melt magma mixing or melt-rock assimilation type mixing or mechanical mixing. However, having determined the likely two end-members above, the mixing is most likely between the KREEP-like melt (with the lowest Sr/Sr* and Eu/Eu*) and the anorthositic (feldspathic) cumulate (the highest Sr/Sr* and Eu/Eu*).

The major element compositions of these samples should provide insights into the true nature of such two-component mixing interpretation. Figure 8 shows Al_2O_3 variation diagrams of major, minor and trace element as well as Sr/Sr* and Eu/Eu* anomaly ratios. We choose Al_2O_3 as the independent variable because it has the largest variation range (6.5–28.7 wt.%) and because it is an effective indicator of the role of plagioclase. Hence, increasing Al_2O_3 means increasing the modal (or normative) plagioclase of the samples, which is consistent with its positive correlation with CaO, i.e., the Ca-rich anorthic plagioclase. The significant inverse correlations of Al_2O_3 and CaO with FeO, MgO and Cr_2O_3 (to some extent with Co and TiO_2) are consistent with the complementary relationship between plagioclase and mafic phases (olivine, pyroxenes plus Fe-Ti oxides and perhaps spinels) in these samples. With the exception of two samples, there is a positive correlation between Al_2O_3 and Ni, suggesting the possible effect of olivine and the anorthositic cumulate may be olivine-poor troctolite although we cannot rule out the effect of sulphides. This suggests the necessity that the lunar community with actual sample access should look into this observation.

From these major and minor element systematics, we can conclude that the lunar samples define a major element compositional continuum from Al_2O_3+CaO poor to Al_2O_3+CaO rich with complementary co-variation of MgO+FeO. This is mathematically a mixing relationship, but may not be so physically. The most logical geological explanation for such a compositional continuum is the mineralogical and lithological variation of the samples associated with their respective petrogenesis histories. For example, those with high Al_2O_3+CaO (low MgO+FeO) are dominantly anorthositic rocks (e.g., characteristic of lunar highland crust) whereas those with low Al_2O_3+CaO (high MgO+FeO) are largely basaltic rocks (e.g., characteristic of lunar mare crust). Not shown, but SiO_2 varies accordingly but in a small range with complications by the KREEP samples (see below). Actually, all the 21 representative samples are from 6 Apollo sites and 3 Luna sites where both types and transitional rocks (or regoliths) exist (see Lucey et al., 2006). Hence, each of the samples represents a rock with its own petrogenesis histories.

The complication added to the above two-end-member lithological mixing is manifested by the data (samples) cluster in CaO-Al_2O_3 and MgO-Al_2O_3 diagrams. The data cluster (guided by the grey bands) in fact includes incompatible element enriched KREEP-like samples with high K_2O, P_2O_5, Ce. These KREEP-like samples also have stronger negative Eu and Sr anomalies (lowest Sr/Sr* and Eu/Eu* ratios), a relationship that is very well expressed by the inverse Th vs. Eu/Eu* correlation in Figure 5. This relationship as discussed above (Figs. 2-5,7) is mathematically a straightforward mixing relationship. The amount of the KREEP-like material involved in the mixing is likely volumetrically small enough not to affect the first-order two end-member lithology trend, but has caused the elevated abundances of incompatible elements in Figure 8. Likewise, the high abundances of incompatible elements and the strongest negative Eu

anomaly of these KREEP-like material (Fig. 2) dictates the Eu and Sr anomalies of the actual samples. That is, the greater KREEP-like component in the sample, the higher incompatible element abundances and the stronger the negative Eu (and Sr) anomalies (the smaller Eu/Eu* and Sr/Sr* ratios) in the sample as manifested in Figures 2-5 and 7. This is also apparent from the lack of Al_2O_3 correlation with Sr/Sr* and Eu/Eu* in Figure 8.

Note that we expect samples with greater modal plagioclase should have higher Al_2O_3 (also high Ca) and thus higher Eu/Eu* and Sr/Sr* ratios, but such correlation is not obvious and samples with low Al_2O_3 and CaO can have higher Eu/Eu* and Sr/Sr* (Fig. 8). Hence, it is the amount of the KREEP-like material component in the sample determines the actual incompatible element abundances as well as the extent of negative Eu and Sr anomalies. In other words, the major element compositional spectrum of lunar samples is controlled by the lithological variation or modal mineralogy variation (i.e., plagioclase rich vs. mafic pyroxene + olivine rich). However, the essentially random addition (mixed in) of KREEP-like material explains the compositional spectrum in terms of abundances of incompatible elements and the size of Eu and Sr anomalies. This is an important observation that we advocate the attention by the lunar scientific community.

5.2.3. Summary on the significance of Eu and Sr anomalies

The parallel REE (Fig. 2) and incompatible element abundance (Fig. 3) patterns suggest that the mixing may be simply addition (or subtraction mathematically). That is, the lunar rocks, whether on the feldspathic highlands or in the basaltic maria, may have been to varying extent randomly contaminated by such KREEP-like melt. This statement implies that the KREEP-like melt is necessarily youngest, younger than the existing highland and mare rocks (Taylor, 1975; Lucey et al., 2006), but this is unknown. It is possible that there is a widespread KREEP-like reservoir (melt or solid rock types in the lunar crust or mantle) that may have been involved in the petrogenesis of lunar highland and mare rocks. The point is that there is no obvious genetic relationship between this KREEP-like component and the lunar rocks in the obtained samples (see Lucey et al., 2006).

Following the foregoing discussion that it is the KREEP-like material and its amount that determine whether the contaminated lunar rocks, especially the highland rocks, can preserve the positive Eu (also Sr) anomaly, *NOT* the intrinsically high Eu/Eu* (also high Sr/Sr*) in the anorthositic highlands as widely thought. Hence, it now becomes straightforward why there is no positive Eu anomaly in the average lunar highlands (see Figs. 5-6 above), as argued previously (O'Hara, 2000, 2004). It follows that all the hypotheses based on the assumption that the lunar highland crust has a positive Eu anomaly must be re-examined (see above).

On the basis of this strong argument alone, we cannot deny the possibility for the presence of the hypothetical LMO, but the very foundation for the LMO hypothesis is no longer valid. As a result, all other aspects of the lunar petrogenesis models built on the LMO hypothesis need reconsideration. Plagioclase rich rocks necessarily have a positive Eu anomaly (also Sr anomaly). Hence, the model lunar highland crust FUpCr constructed based on lunar meteorites with abundant plagioclase (see Lucey et al., 2006) are expected to have very high positive Eu and Sr anomalies (Fig. 7). However, meteorites are physically strong coherent rock masses selectively sampled and landed on the Earth by nature and cannot represent the lunar surface rock composition because the lunar surface materials are largely regoliths. Materials that can better represent lunar surface rocks are returned samples, but only the remote sensing data averaging over large areas are statistically more representative of the lunar crustal rocks if and only if the surface regoliths indeed represent the subsurface rocks and the crustal mass.

5.3. More insights from remote sensing data and lunar surface compositions

The greater the plagioclase content of the rocks, the lower must be the FeO and TiO_2 contents (see Fig. 8). The greater the feldspar content of the rocks, the higher will be the Al/Si ratio, their Al_2O_3 and CaO contents. All these parameters were measured by the Gamma Ray Spectrometer aboard Lunar Prospector (LP-GRS). These parameters, individually and in combination, demonstrate the scarcity of significant areas of the lunar surface with feldspar contents as low as those of the *assumed* primary magmas. They also provide evidence of a negative (vs, positive) Eu anomaly in the average lunar highland crust because the dominant lunar surface has Th > 0.7 ppm (see Figs. 5,6) .

5.3.1. Ti: Mare regolith TiO_2 contents are much lower than in hand specimens from Apollo 11 and 17. They may never exceed those of experimentally produced plagioclase- and ilmenite-saturated basaltic liquids at low pressure

The story begins with a simple test of the validity of the first alleged lunar primary magmas. The first and last of the six Apollo landings returned high-Ti basalts, with regoliths containing 8-9 wt% TiO_2. Hand specimens from these two sites contained 12-14% TiO_2; they were the first samples to be asserted to be 'primary' magma compositions, and they are required to represent the average compositions of flood basalt flows (see O'Hara, 2000).

The Lunar Prospector data, however, indicate that there is relatively little high Ti regolith material on the mare surfaces (Fig. 9), very little with TiO_2 contents higher than the low-pressure plagioclase-saturated liquids (9-11% TiO_2) and almost none with TiO_2 >12%. *NVOTM* (2006) emphasises the case that there are few areas of the lunar surface, and no returned soils, whose TiO_2 contents match those of the hand specimens or exceed those of the cotectic liquids. This observation is inconsistent with the hypothesis that the Apollo 11 and 17 hand specimens are quenched samples of volumetrically significant primary magmas, or that they represent the average composition of extensive flood basalt magmas. Figure 9 demonstrates that the returned samples or meteorites do not represent the lunar surface materials (rocks or regoliths), but localized Fe-Ti rich materials due to sampling "bias" compared to lunar compositions revealed by the global remote sensing data (O'Hara 2000, 2001a). The observation can be explained as a straightforward consequence of fractional crystallization of basaltic parental magmas at a late stage (basaltic-andesite stage; Niu et al., 2002; Niu, 2005; Lustrino, 2006; Stone and Niu, 2009), when Fe-Ti oxides reach the liquidus. For example, the well-known Panzhihua Fe-V-Ti ore deposit (ilmenite-pseudobrookite-titanomagnetite assemblage) in Sichuan, China, is of such origin (Niu, 2005) with the Ti reserve ranked the very top in the world; the parental basaltic magmas are part of the ~250 Ma large igneous province. Hence, we consider the Ti-rich hand specimens from Apollo 11 and 17 as cumulus-enriched samples, whose biased sampling is natural as they differ uniquely from the rest regolith materials.

5.3.2. Fe: Very little of the mare surface has FeO as high as the alleged primary basalt magmas (represented by the hand specimen basalts).

Low-Ti basalts form much greater areas of the mare surfaces. Hand specimen samples recovered by Apollo 12 and 15 were in their turn asserted to represent the compositions of primary magmas. These samples, containing 19-22% FeO, would again be required in the 'established' model to provide the average compositions of extensive flood basalts on the mare surfaces. Figure 10 demonstrates that this is not true – there is a peak in the frequency of values at 13-19 wt% FeO, most likely resulting from earlier plagioclase (vs. pyroxene) crystallization/removal from the

parental melts. If the picritic glass beads are advanced as the primary magmas, their contribution to the surface area is evidently small. Again, it can be the cumulus-rich materials of straightforward consequence of fractional crystallization of basaltic parental magmas as above for Ti-rich specimens (see above in the context of Ti-Fe enrichment and mineralization). That is, they are cumulate from highly evolved basaltic melts at low pressure rather than primary magmas (see O'Hara, 2000).

5.3.3. Insights from Al_2O_3 vs. FeO space

There is strong negative correlation between Al_2O_3 and FeO (Fig. 8) in the samples returned by the Apollo 15 mission (Fig. 11). This high resolution sampling of a low-Ti basalt surface confirms the generally feldspathic nature of the average target rock on the mare surfaces. A similar plot (Fig. 12) of LP-GRS results illustrates the global paucity of surfaces as rich in iron and poor in alumina as the putative low-Ti basalt primary magmas, or of surfaces feldspathic enough to be likely to display positive Eu anomalies. This correlation results from a data closure because of the mineralogical complementarity (Fig. 8), i.e., Al_2O_3+CaO rich anorthositic rocks vs. MgO+FeO rich basaltic rocks. But this does not weaken the argument here, i.e., returned samples do not represent the compositions of the lunar surface materials (rocks or regoliths) and the surface materials, if they are of magmatic origin, are low-pressure cotectic compositions with plagioclase as a liquidus phase (e.g., gabbroic fractionation with gabbro, gabbro-norite and even Fe-Ti oxide cumulate).

5.3.4. Insights from Al_2O_3 vs. CaO space

Al_2O_3 and CaO concentrations are now available for the whole lunar surface as shown in Figure 13. The paucity of compositions close to that of pure anorthite, where a throng positive Eu anomaly is guaranteed (Fig. 7), is evident. Equally conspicuous in Figure 13 is the paucity of compositions overlapping the field of alleged alumina-poor, iron oxide rich primary magmas in the Maria (two fields shaded green at lower left).

5.3.5. Insights from Al/Si Ratio: Hand specimens do not represent average erupted basalt

O'Hara (2000) demonstrated that Al/Si ratios in remotely sensed lunar surface materials suggest that there is no positive Eu anomaly in the average lunar highlands (see above) and that average mare basalt compositions are significantly more feldspathic than the hand specimens. These data were, however, restricted to the equatorial belt and were obtained by XRF with low penetration. The Al and Si concentrations by the LP-GRS are now available for the whole lunar surface. Figure 14 confirms the earlier values of Al/Si <0.27 required to match any surface areas corresponding to the putative primary magma compositions. Figure 14 also displays peak concentrations close to Al/Si ~0.65, with a low frequency of values above 0.8 at which a positive Eu anomaly can be expected. The profound implications of these results are not discussed in *NVOTM* (2006)

6. How representative are returned samples, meteorites and LP-GRS data of the lunar surface regoliths and lunar crust?

Korotev et al. (2003) endeavoured to estimate the average composition of lunar surface and crustal compositions using highland-derived meteorites. The FUpCr (Figs. 3, 7) is the best estimate for typical feldspathic lunar crust of upper few kilometers, which is very similar to their estimated surface composition of a fee meters (see Table 2.6 given by Lucey et al., 2006). This is a highly commended effort. However, as discussed above, meteorites are coherent solid masses (vs.

regoliths) selectively sampled and landed on Earth by nature, and they thus cannot represent the mean composition of the surface nor subsurface compositions as revealed by the global LP-GRS compositions. The mission-returned samples and meteorites are important, but are biased by the nature of sampling. However, the LP-GRS data give representative mean compositions of the lunar surface material although further effort is needed to obtain truly representative bulk lunar crust compositions by, hopefully, sampling the deep crust. Our goal is thus to obtain bulk crustal compositions towards much better improved understanding of the lunar magmatism and petrogenesis.

6.1. Deeper sampling of highland terrain by central peaks of large craters

Impact crater central peaks offer a unique opportunity to remotely examine material that was once deep in the crust (Cahill et al., 2009). The majority of peaks analyzed had compositions similar to Mg-suite rocks of the lunar sample collection, independent of lunar terrain, contrary to one of the principal conclusions by Korotev (2005) based on lunar highland meteorites. The results also indicate that plagioclase-rich materials which might carry a positive Eu anomaly become no more abundant with depth than they are on the surface.

6.1.1. The Lunar mare meteorites

Mare soils, which can contain only a small highland contribution, are much more aluminous (feldspathic) than the hand specimens, which cannot therefore represent the average target rock at the Apollo 11, 12, 15 or 17 sites – or elsewhere on the Maria (see O'Hara, 2000). Randomly sampled basalt lithic fragments are slightly more feldspathic than the plagioclase saturated cotectic liquids, leading to the conclusion that the erupted magmas would have crystallised plagioclase slightly before pyroxene (see O'Hara, 2000). These propositions are examined further in the context of low-Ti basalts sampled at the Apollo 15 site, and by the lunar mare meteorites (O'Hara, 2000).

The lunar meteorites may have all been launched relatively recently by a possibly restricted number of individual impact events (see O'Hara, 2000). Scarcity of the material leads to chemical analyses which are not guaranteed to represent the average composition of each meteorite. They represent a randomly chosen suite of real, but necessarily close-to-average samples comparable with and supplementary to the Apollo and Luna sampling in this respect. Korotev (2005) listed 36 launch events, 21 of which (total mass ~0.009 metric tonnes) were highland-derived, 5 (total mass ~0.003 metric tonnes) purely mare-derived and 10 (~0.015 metric tonnes) of mixed target character. Lunar mare meteorites are thus 'qualified' random samples of mare near-surface materials. All these samples are low or very low in Ti. Most are as feldspathic as, or more feldspathic than, the low-Ti plagioclase-saturated cotectic liquids. Ophitic or sub-ophitic texture in several samples indicates early nucleation of plagioclase. Coarse grain size indicates slow cooling, favouring shallow crustal level differentiation. LAP 02205 is calculated to be compositionally cotectic for plagioclase, pyroxene and olivine, indicating that this is NOT a primary magma, but one that was undergoing low-pressure gabbroic fractionation. The Al_2O_3, SiO_2 contents and Al/Si ratio of lunar mare meteorites are compared in Figure 15 with data for the most extensively studied low-Ti basalt site (Apollo 15).

The data from the lunar mare meteorites support the availability of target basalts significantly more feldspathic than the hand specimens, i.e., they have plagioclase on the liquidus equilibrium under low-pressure condition and are NOT primary magmas.

6.1.2. The Lunar highland meteorites

Korotev (2005) reported 21 highland derived meteorites, all the analysed samples displaying a positive Eu anomaly with Th <0.5 ppm, and Al_2O_3 = 26-33%. This suite displays low total REE concentrations nearer ~10x chondritic, rather than the nearer 50x chondritic values found in the returned Apollo sample suite. *NVOTM* (2006) prefers this average as an estimate of initial highland crust composition. However, as elaborated above, meteorites are physically coherent masses selectively sampled and landed on the Earth and cannot represent average lunar highland compositions of loose regoliths, which is clear in Figure 13. This straightforward and logical consideration must be exercised in constructing model average compositions of lunar crust. This highly feldspathic rock type could be volumetrically abundant in favour of the LMO hypothesis, but the positive Eu anomaly is overshadowed by the KREEP-like material enriched in non-Eu REEs and other incompatible elements with strong negative Eu and Sr anomalies (Figs 7-8).

6.1.3. Mare Imbrium in detail

Bugiolacchi and Guest (2008) studied the basalts of Mare Imbrium. They found, from Clementine and Lunar Orbiter data, a complex suite of low to high-Ti basaltic lava units infilling the Mare Imbrium basin, mainly over an 800 Myr interval, but with evidence of an almost uninterrupted record of exposed lava flows spanning 1.4 Gyrs within the basin. Estimated emplacement ages between 3.3 and 2.5 Ga were inferred for 75% of the sampled area. 18% of the sampled surface in mare Imbrium contained FeO between 14 and 16 wt%, 55% had FeO 16-18%, and 27% had 18–20 wt% FeO. No significant areas were identified with FeO ~20-24% characteristic of putative primary low-Ti basalt magmas (basalt hand specimens and picritic glass beads) from Apollo 12 or 15.

About 37% of mare Imbrium basalts contained 1–3 wt% TiO_2; two other areas, each of about 25% of the surface, showed TiO_2 values between 3–5 and 7–9 wt%. No significant areas were identified with TiO_2 values between 10–14 wt% characteristic of putative primary high-Ti basalt magmas (basalt hand specimens and picritic glass beads) from Apollo 11 or 17. A correlation between the FeO and TiO_2 was identified, in particular for materials with TiO_2 and FeO content above 5 and 17 wt%, respectively, suggesting a related petrogenesis and evolution, which is expected as cumulus-rich material as a straightforward consequence of basaltic magma evolution as manifested by the magmatic Fe-Ti-V deposits discussed above (Niu, 2005; Niu et al., 2002; Lustrino, 2006; Stone and Niu, 2009).

Three major periods of mare infill are exposed in the Imbrium basin and in each period, igneous petrogenesis generally evolved through time towards more TiO_2 and FeO-rich liquids. This evidence is easily reconciled with the view that parental liquids were low in Ti and residual liquids increased in Ti content as fractionation of gabbros in near-surface magma chambers advanced (Niu, 2005; Stone and Niu, 2009). They are not primary magmas for reasons elaborated above. Bugiolacchi and Guest (2008) concluded in favour of a primary magma model without discussion but asserting "…the geographical distribution of basalt types classified according to their titanium abundances is consistent with the presently accepted models of crystallization of the inferred lunar magma ocean."

6.2. Are we trying to answer the right question?

Average mare basalt regolith compositions are more feldspathic, in terms of Al_2O_3 content (Fig. 16), than the low pressure plagioclase-saturated cotectic liquid compositions and much more feldspathic than the hand specimens or picritic glass beads. Average highland regolith

compositions are less feldspathic than the anorthositic materials which carry significant positive Eu anomalies. Thus far, it might be argued, there has been too much manpower devoted to explaining the extremes of anorthite-rich and anorthite-poor materials which flank that broad central distribution.

The fundamental petrogenetic problem of the lunar crust is to explain the super-abundance of compositions ranging from feldspathic basalts to highly feldspathic noritic materials – which are accompanied by a negative Eu anomaly in the average bulk crust composition (Fig 17). It is the wide range of $Mg^{\#}$ in the densely populated region of the plots at low MgO, FeO, and high Al_2O_3 which suggests melt evolution along some cotectic saturated with plagioclase and one or more of olivine and the pyroxenes. Are there conditions under which such a cotectic exists? We must also keep in mind that most of these surface regolith materials do not represent melt composition, but mixtures of melt and cumulate rocks.

7. Origin of the lunar crust

At present, we can only discuss the petrogenesis of the lunar crust by assuming that the bulk lunar crust is of magmatic origin and that other processes are negligibly unimportant, including extra-terrestrial impact contribution to the mass and lunar surface composition.

7.1. Average lunar crust as a partial melt

The average composition of the whole surface of the lunar crust from LP-GRS results contains 42.33 wt% SiO_2, 1.21 wt% TiO_2, 24.46 wt% Al_2O_3, 7.72 wt% FeO, 8.75 wt % MgO, 15.17 wt% CaO, and $Mg^{\#}$ = 69.2 (assuming 90% of total Fe as Fe^{2+}), close to the value expected for melts in equilibrium with mantle olivine of Fo_{90} (assuming an Earth-like peridotite mantle). Average Th is 2.27 ppm, consistent with an overall significant negative Eu anomaly (Fig. 5) in the crust and a positive Eu anomaly in the lunar mantle as predicted in Earth's mantle (Niu and O'Hara, 2009) although this assertion needs verification given different lunar and Earth rock trace element systematics (Fig. 3). Assuming a broadly peridotitic bulk composition for the primitive lunar mantle, what processes of melting could extract such an Al_2O_3-rich (consistent with a high feldspathic composition) and high $Mg^{\#}$ material (Fig. 17)? on a global large scale?

7.2. What mass fraction of which partial melt?

If the lunar crust has been derived from the whole Moon, an average of 5-8 % melting is required. Approximately three times greater required if the crust is extracted from the outer 250 km only. O'Hara (2000) considered the classic model of "spherical zone refining during accretion" (SZRA) is a good model to explore. Walker (1983) advocated a gradual and piecemeal model for lunar magmatism and crust formation without the need of a magma ocean. *NVOTM* (2006) insists that any calculations on the extent of melting using SZRA type of process (vs. LMO) based on Al_2O_3 or Th budgets would be premature.

7.3. Simple dry-moon model with the development of a magma ocean

First, let us assume a model of accretion of a volatile-depleted Moon *followed by* the partial melting event. The peridotite mantle of such a body, once accreted, lies at pressures and temperatures within the plagioclase-lherzolite mineral facies to a depth of ~250 km; within the spinel-lherzolite facies from ~250 km to 400 km (at 700-1000 ℃), or up to 650 km at near solidus temperatures; and within the garnet-lherzolite facies at even greater depths. The lowest pressure partial melting products differ significantly from the average lunar highland crust and diverge further as pressure

increases. Melt compositions produced by low mass fractions of partial melting at the dry solidus of such a mantle would be basaltic close to the surface but would become more feldspathic, increasingly troctolitic but also increasingly olivine-normative, as depth increased to ~250 km, and thereafter increasingly picritic (Herzberg and O'Hara 1998, 2002, O'Hara and Herzberg 2002). More advanced partial melting at any pressure increases the mafic character of the melt products.

A variant on the above would be essentially the LMO model, requiring a much greater energy input into enthalpy of melting in order to "process" the planet, and requiring a positive Eu anomaly in the average crust composition that is globally not observed (Figs. 5-7). The non-dry Moon nature, as inferred experimentally (O'Hara et al., 1970; O'Hara, 1972, 2000) and confirmed recently by measured H_2O in lunar glasses and minerals (Saal et al., 2008; Liu et al., 2012; Hui et al., 2013), denies the dry moon assumption. The presence of volatiles in abundance in lunar minerals and glasses further suggests that the high energy impact may not have happened to Earth before, further weakens the LMO hypothesis as the invoked planetary impact would have removed all (or almost all) the volatiles.

7.4. Simple undepleted-moon model

Let us now assume a model of accretion of a volatile-containing Moon from material close to carbonaceous chondrite plus ices in average composition, with significant amounts of alkalis, water and carbon dioxide retained at depth. The critical factor at the start of melting is the presence of the fourth, alumina-rich, phase in addition to olivine and two pyroxenes (O'Hara 1965, 1968) plus the possible stability of amphibole, mica or carbonate. That fourth phase (feldspar, spinel or garnet) was present in the experiments in the systems CMASH. CMASNH and CMASKH surveyed in the fine-print section below.

In the presence of water and alkalis, the silica and feldspar contents of peridotite partial melts are increased at low to moderate pressures, but tend towards silica-undersaturated, not highly feldspathic compositions at high pressures. Reducing the activity of H_2O in the system by the presence of CO_2 at low overall pressures offers a solution (see fine-print section) but the volume of a fully accreted Moon within the requisite low pressure regime may be inadequate to provide the mass of the lunar crust.

Fine-print

The partial melting of simplified forsterite + 2 pyroxene + aluminosilicate assemblages resembling wet plagioclase-, spinel-, and garnet-lherzolites in the system CMASH was encountered without the intervention of hydrous crystalline phases at 0.5 GPa, 1010 ℃ by Ford and O'Hara (1972) on the enstatite-grossular join; at 980-1000 ℃, 1 GPa by Yoder and Chinner (1960) on the pyrope-grossular join, by Yoder (1971) on the diopside-pyrope join, and by Ford (1974) on the enstatite-grossular join; at 1000 ℃. *Liquids produced are quartz-normative and of decreased CaO/Al_2O_3 ratio.*

Partial melting of simplified soda-containing forsterite + 2 pyroxene + aluminosilicate assemblages resembling wet plagioclase-, spinel-, and garnet-lherzolites in the system CMASNH was encountered, (just) without the intervention of hydrous crystalline phases at 960 ℃, 0.5 GPa by Cawthorn (1976).

At higher pressures Kushiro (1972) found a quartz-normative partial melt of a garnet-lherzolite + vapour assemblage in CMASNH at 1075 ℃, 2.5 GPa. The composition used had a CAS:NAS_4 ratio of 4.9 and Kushiro (1972) predicted that at lower ratios the water-saturated partial melt might be nepheline-normative. This was confirmed by Howells (1975, 1978) who obtained nepheline-normative liquids containing 23% water, in equilibrium with garnet-lherzolite at 1075 ℃, 2.5 GPa in the presence of a water-rich vapour, and further nepheline-normative liquids in the vapour-free equilibrium up to 1200 ℃, 11% water. The liquids concerned contain more than 3-4% Na_2O wt%.

Partial melting of simplified potash-bearing forsterite + 2 pyroxene + aluminosilicate + phlogopite + vapour assemblages resembling wet phlogopite-bearing spinel-, and garnet-lherzolites in the system CMASKH was studied by Bravo and O'Hara (1975) who found the equilibrium at 980 ℃, 1.5 GPa with the

production of a quartz-normative andesitic liquid with about 6% orthoclase in the norm; and at 1080 ℃, 3 GPa with the production of a liquid rich in orthoclase, hypersthene and olivine in the norm.

Addition of carbon dioxide to model lherzolite and aluminous lherzolite in $CMS.CO_2$ and $CMAS.CO_2$ produces dramatic effects on temperatures and compositions of liquids produced at pressures greater than 2.9 GPa (e.g., Wyllie, 1977; Wyllie et al., 1983; Dalton and Presnall, 1998) but this pressure corresponds to a depth of about 700 km in the Moon. At lower pressures solubility of carbon dioxide in the melts is low and its principal effect will be to reduce the activity of water in the fluid phases. Partial melting at temperatures below 1000 ℃ will occur at pressures above 0.2 GPa. Partial melts produced in the pressure range below 0.5 GPa might be expected to be intermediate in composition between feldspathic basalt or troctolite (dry end) and silica-saturated andesitic (wet end), i.e. not unlike the average composition of the lunar crust – and they could be produced in equilibrium with a plagioclase-bearing residuum in this shallow depth range, thereby imparting negative Eu and Sr anomalies to the melt.

Below 1 GPa, in the plagioclase-lherzolite field, partial melting of plagioclase-bearing lherzolite might plausibly yield liquids with overall negative Eu anomalies. In the dry systems those liquids would tend towards troctolitic and noritic compositions too poor in silica to approach the lunar crust bulk composition. The low solubility of carbon dioxide in silicate melts in this region suggests that its effects on the silicate melt composition will be subordinate, beyond reducing the activity of water, to those produced by the presence of water, which are to reinforce the shift towards decrease in CaO/Al_2O_3 ratio, i.e. towards increase in potential feldspar, while counteracting the shift towards increased olivine in the CIPW norm. The effects on the CIPW norm of sodium loss by selective vaporisation together with the water and carbon gases may be substantial until the growing body is larger than the Moon and should not be overlooked.

7.5. A possibility worth pursuing

Involvement of H_2O, CO_2 and alkalis in a mixed relatively volatile-rich proto-Moon undergoing the classical "spherical zone refining during accretion" (SZRA) process might lead to products which will bear further examination in the lunar context (e.g. O'Hara et al 1970a, b, Ford et al 1972, O'Hara 2000). Such a model might apply equally to the accretion and primary crust formation on Mercury, Mars, Venus and the Earth. In this context, the views of Lowman and Yang (2005) are relevant. They present photo-geologic evidence from the Apollo 15, 16 and 17 missions of a pervasive layered structure in the highlands. The number, thickness, and extent of these layers implies that they are lava flows, not ejecta blankets or intrusive features and they are interpreted as the upper part of the earliest lunar crust, possibly forming a layer tens of kilometers thick representing a global crust formed by eruptions of high-Al_2O_3 basalt in the first few hundred million years of the Moon's history. We suggest that the SZRA is a worthwhile alternative to examine for the lunar magmatism and crustal formation (see O'Hara, 2000).

7.6. Consequences for Al_2O_3 distribution within the Moon and other planets.

Extraction of a feldspathic planetary crust by partial melting during SZRA (vs. LMO) has major implications for the distribution of Al_2O_3 within any planet. SZRA can be more efficient in concentrating Al_2O_3 into the crust. Both processes create an upper mantle of residual peridotite whose pyroxenes are saturated with alumina at the conditions of equilibration. At 1 GPa the temperature is likely to be ~1000 ℃ in the former process, ~1250 ℃ in the latter. At these conditions SZRA might lock up in the mantle pyroxenes ~3 wt% Al_2O_3 less than expected in the LMO case. This would imply ~2 wt%, but possibly much less, *throughout* the resultant mantle as opposed to ~3.5 wt% in a *restricted outer zone* where plagioclase saturation had been reached - an order-of-size estimate based on Gasparik (2000) and about 50% pyroxene in the assumed mantle. The difference might be greater because the SZRA model is a partial melting process which requires only that the pyroxene be present and alumina-saturated. The LMO model is a fractional crystallisation process which constrains the ratio of olivine to pyroxene in the precipitated mantle.

Given the unlikeliness of the LMO, we suggest that the classical SZRA is a physically and chemically robust mechanism to be explored in the future.

8. Summary and preliminary conclusions

Following the above thorough discussion, data analysis and logical reasoning, we can suggest and conclude as follows.

1. Despite the valuable achievements over the past decades on the petrogenesis of lunar rocks, some basic issues recognized in early days remain poorly addressed. These issues need resolving before we proceed on *what's left to learn*.
2. These basic issues are actually unsatisfactory assumptions built in the established models, e.g., *dry Moon*, *primary lunar magmas*, *plagioclase flotation* and the central paradigm of the global *lunar magma ocean* (LMO).
3. The senior author emphasized the likely importance of volatilization during lunar magma emplacement (O'Hara et al., 1970; O'Hara, 1972), suggesting that the Moon must have been a water-bearing (if not water rich) planetary body, but this rock/mineral phase equilibria-based reasoning has been entirely neglected by the lunar research community until recent determination of water in lunar glasses and minerals (Saal et al., 2008; Hui et al., 2013). Among other implications, degassing (e.g., water loss) further facilitates plagioclase crystallization at low pressure (e.g., Yoder, 1965), which explains plagioclase in abundance in lunar crust, not only in the anorthositic lunar highlands, but also in the Maria.
4. Importantly, if the Moon has indeed had abundant volatiles in its early histories, or still so in its deep interior, then this would question the *planetary impact origin* for the Moon and the *LMO hypothesis*, because the original volatiles would have been largely or entirely evaporated. The actual presence of volatiles (in particular water) in the Moon further questions the *dry moon hypothesis*.
5. The *LMO* was required for *plagioclase flotation* as the very origin of the anorthositic lunar highland crust with a strong positive Eu anomaly and the cumulate origin of the lunar mantle as the sources for mare basalts with inherited negative Eu anomaly. Alternative models (vs. LMO) are needed for lunar petrogenesis.
6. As plagioclase is preferentially enriched in Eu relative to other rare earth elements, plagioclase-rich rocks like anorthosite, troctolite, gabbro and gabbro norite are enriched in Eu with a positive Eu anomaly (i.e., Eu/Eu* >1). Indeed, returned lunar highland rocks and meteorites enriched in plagioclase all have a strong positive Eu anomaly. As a result, the lunar highlands have been assumed to be characterized by a positive Eu anomaly, which in turn has been considered as the most convincing evidence that underpins the *plagioclase flotation* hypothesis. However, the lack of a positive Eu anomaly in average lunar rocks in general and lunar highlands in particular weakens the foundation for *plagioclase flotation* hypothesis and further refute the LMO hypotheses (Figs. 5-6).
7. In this context, it should be noted that the lack of a positive Eu anomaly in the lunar highlands does not negate the anorthositic bulk compositions in terms of the mineralogy and lithology, but does mean that we cannot use the assumed positive Eu anomaly as evidence for the preferred models.
8. Lunar meteorites are valuable materials for understanding the composition and petrogenesis of the Moon. However, they are physically coherent material masses selectively sampled and landed on Earth by nature and cannot represent true composition

of the lunar surface rocks and regoliths. The mission-returned samples are similarly valuable, but they do not represent the mean composition of the vast lunar surfaces and lunar crust. Nevertheless, the feldspathic lunar surface composition is acquired by the LP-GRS data (Figs. 6,9-17) and such data are more representative in discussing the first-order processes on lunar petrogenesis in particular and crustal formation in general.

9. If lunar surface samples compositionally represent magmatic rocks, then compared to the Earth magmatic rocks, the lunar rocks are highly depleted in Na (Na_2O <0.8 wt%; Fig. 8) and other alkali elements K, Rb Cs as well as P (Fig. 3). These are all moderately volatile elements that could have been partially lost during accretion in the very early histories of the Moon. Magma Oceans have been hypothesized for both the Moon and Earth, so the reasoned lunar alkali volatilization may have nothing to do with the hypothesized Magma Ocean processes as otherwise the Earth would have also lost these elements, but it does not (Fig. 3). Therefore, further research is necessary to understand the compositional contrast between Earth and Moon rocks seen in Figure 3.
10. The feldspathic lunar surface composition is consistent with the important role of plagioclase in the lunar petrogenesis (Figs. 8, 11-17). This is further manifested by the correlated variation of Eu/Eu* and Sr/Sr*(Fig. 7) because plagioclase is the only mineral that can effectively and simultaneously fractionate these two elements from otherwise similarly incompatible elements during plagioclase crystallization. However, different from the correlated Eu/Eu*-Sr/Sr* trend defined by Earth rocks such as mid-ocean ridge basalts resulting from cooling-induced fractional crystallization (Fig. 7), the linear Eu/Eu*-Sr/Sr* trend defined by the lunar rocks is a mixing trend between rocks/melts with Eu/Eu* and Sr/Sr* >1 such as the anorthositic lunar meteorite, and rocks/ melts with Eu/Eu* and Sr/Sr* <<1, such incompatible element highly enriched KREEP-like materials (Figs. 2-5 and 7 read in combination).
11. Despite the globally feldspathic lunar surface composition (Figs. 11-17), the lunar surface rocks do define a compositional continuum from the more feldspathic (relatively Al_2O_3+CaO rich and FeO+MgO poor) to less feldspathic (relatively Al_2O_3+CaO poor and FeO+MgO rich) compositions (Fig. 8). The globally mean negative Eu and Sr anomalies for both highland and Mare surface rocks (Figs. 3,5-8) suggest that the KREEP-like materials may be widespread throughout the lunar surface although they may be areally (or volumetrically) less significant (?). The lack of Al_2O_3+CaO correlation with Sr/Sr* and Eu/Eu* manifest that although the plagioclase-rich rocks (with high Al_2O_3+CaO) are expected to have large positive Eu and Sr anomalies, the actual size of the anomalies is largely controlled by the amount of KREEP-like mixing component. The mixing is more consistent with a "simple mechanical" mixing, rather than magma source mixing or melt-rock assimilation type mixing as manifested by the simple linear relationship on element-element plots (Figs. 2-4) and simple curvilinear relationship on element-ratio plot (Fig. 5). On Sr/Sr*-Eu/Eu* plot (Fig. 7), the glass beads lie on the linear mixing trend defined by other lunar samples. It is possible that these glass beads may actually be melt drops of impact-induced in situ melting origin.
12. The origin of KREEP and KREEP-like materials is consistent with prior advanced extent of fractional crystallization with elevated concentrations of highly incompatible elements and extreme depletion in Eu and Sr. It should be noted that such enrichment process is not a simple equilibrium crystallization (inadequate) but repeated enrichments effectively through fractional crystallization equivalent to processes "*in an advancing, periodically*

replenished, periodically tapped, continuously fractionated magma chamber" (O'Hara, 1977; O'Hara and Mathews, 1980).

13. The mare basalts and basaltic rocks do not represent primary magmas, but products after varying extents of low-pressure gabbroic fractionation (Plagioclase-pyroxene-olivine cotectic). Hence, their negative Eu (also Sr) anomalies are likely caused by plagioclase crystallization rather than inherited form the mantle source. We cannot rule out the possibility that the lunar mantle sources may have positive Eu and Sr anomalies as is the case in the Earth's mantle (Niu and O'Hara, 2009).
14. Despite the feldspathic mean composition of the lunar surfaces, high FeO and high TiO_2 materials have been sampled (both meteorites and mission-returned) and observed from the LP-GRS data (Figs. 9-12). High FeO and TiO_2 melts are rare on Earth but do exist in highly evolved MORB melt (up to 19 wt% FeO and 4.0 wt% TiO_2) as a result of advanced extent of fractional crystallization before Fe-Ti oxides on the liquidus under low fO_2 conditions (Fornari et al., 1983). Many large Fe-Ti-V ore deposits are actually of cumulate origin genetically associated with such highly evolved basaltic melts and there is also the possibility that such deposits can directly precipitated from Fe-Ti melt segregated from such highly evolved basaltic melt (at basalt-andesite stage; Niu, 2005; Niu et al., 2002; Stone and Niu, 2009). Hence, the Fe and Ti rich materials on the Moon may be of similar origin.
15. We suggest that more effort is needed to seriously test or reconsider the LMO hypothesis and alternative processes such as spherical zone refining accretion (SZRA) and other processes need to be considered towards an objective and genuine understanding of the origin and evolution of the Moon and lunar petrogenesis.

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Figure Captions:

Figure 1.

The lunar magma ocean model (LMO) explained in rare earth element (REE) pattern diagram. Plagioclase flotation in the LMO produced the anorthositic lunar highlands with Eu/Eu* >1 and the mafic cumulate sank in the mantle as the source for the mare basalts with a complementary Eu/Eu* <1. The KREEP represents widespread yet volumetrically insignificant and highly evolved (with significant plagioclase removal) residual melts enriched in K, REEs, P (hence, KREEP) and other incompatible elements. (After S.R. Taylor, 1982).

Figure 2.

Left two panels are chondrite-normalized REE diagrams for representative retuned samples from both Maria and lunar highlands using average highland ferroan anorthosite (FAN) and KREEP as reference end-members. For Maria samples: HT = high Ti basalts, LT = low Ti basalts, and VLT = very low Ti basalts. For highlands samples: FMB = Low-K Fra Mauro Basalt, RTH = Ross Taylor Highland average, TR = Troctolite, AG = Anorthositic gabbro. FUpCr = Feldspathic Upper crust, GA = Gabbroic Anorthosite (Data from Taylor, 1975; Papike et al., 1997; Lucey et al., 2006; Saal et al., 2008). The right hand-side panel shows a series of samples from the lunar highlands with hugely varying abundances of REEs (but Eu) (Lunar Sourcebook, 1991), in which there is an apparent inverse correlation of Eu/Eu* ($=2Eu_N/]Sm_N+Gd_N]$) values with increasing non-Eu REE (and all other incompatible element) abundances.

Figure 3.

Earth's primitive-mantle (Sun and McDonough, 1989) normalized incompatible element abundances of comprehensively representative lunar surface samples (soils/regoliths) on Apollo and Luna sites summarized by Lucey et al. (2006), plus the model composition of feldspathic upper highland crust (FUpCr; Lucey et al., 2006). The Earth's average OIB (ocean island basalts), enriched E-type and depleted N-type MORB (mid-ocean ridge basalts) and bulk CC (continental crust) are plotted for comparison. If the lunar surface samples (SiO_2, 38.7 – 47.7 wt%; TiO_2, 0.49 – 9.6wt%; Al_2O_3, 6.5 – 28.7wt%; FeO, 4.14 – 22.3wt%; MgO, 4.6 – 14.5wt%; CaO, 7.5 – 16.4wt%)

do represent magmatic products, then the lunar magmatism differs significantly from magmatism on the Earth (see text for discussion).

Figure 4.

Linearly correlated variation of REEs such as Ce with other incompatible element (Ba, Rb, Cs, Th, U, Nb, K, Zr, Y and P) abundances of lunar regolith samples of Apollo and Luna sites (Lucey et al., 2006). In Earth magmatism, these elements have varying incompatibility (i.e., increasing incompatibility from right to left in Fig. 3), but these same elements of the lunar sample show essentially the same incompatibility with varying levels of enrichments (Fig. 3) and parallel correlated linear variations. This rules out lunar mantle source heterogeneity and varying extent of melting as the cause (based on Fig. 3 alone), but is consistent with varying extent of fractional crystallization and/or with simple mixing of enriched KREEP-like melt and depleted anorthositic cumulate.

Figure 5.

From the left panel of Figure 2 on the lunar highland samples, it is clear that there is an apparent correlation between incompatible element abundances (like non-Eu REEs and all other incompatible elements shown in Fig. 4) with Eu anomalies. This is in fact seen in samples from both the highlands and Maria as manifested by the curved inverse correlation of Th abundances (ppm) and Eu anomalies; both regolith samples from all the Apollo sites and Luna sites (Lucey et al., 2006) represented by the red dots and highland rocks chips (Table 8.2 in Lunar Source book, 1991) represented by the thick blue regression line. Both give a very similar power-law fitting. That is, on average lunar samples (from both Maria and highlands) with Th > ~ 0.7 ppm have a negative Eu anomaly (i.e., Eu/Eu* < 1) and samples with Th < ~ 0.7 ppm have a positive Eu anomaly (i.e., Eu/Eu* > 1). This is conceptually the same as the division of Th ~ 0.91 ppm as suggested by Korotev and Haskin (1998). It is thus clear that most lunar surface samples retuned and studied have Eu/Eu* < 1. If Eu/Eu* > 1 indeed points to lunar anorthositic crust genetically resulting from plagioclase floatation in the LMO, then such crust is areally (if not volumetrically) rather insignificant, which may not be true (see text for discussion).

Figure 6.

Frequency of Th concentrations from gamma ray spectrometry (GRS) for the whole lunar surface (see Elphic et al. 2000b, Fig 7). Frequency of concentrations peaks close to 1.5 ppm, with a low frequency of values below 0.8 ppm at which a positive Eu anomaly can be expected. This histogram is based on 259200 averages of the top ~ 0.3m of regolith in areas of ~225 km ², or ~ 108 metric tons of regolith averaged in each observation, approximately 20 trillion metric tons in total.

Figure 7.

This compares the lunar samples (coloured dots) and MORB samples (small solid black circles) in Eu/Eu* vs. Sr/Sr* (=$2Sr_N/[Pr_N+Nd_N]$) space. The MORB trend (Niu and O'Hara, 2009) simply results from cooling induced fractional crystallization. That is, with cooling and decreasing MgO, MORB Eu/Eu* and Sr/Sr* decrease linearly because of plagioclase crystallization. Plagioclase is the single mineral that can effectively and simultaneously fractionate both Eu and Sr from otherwise similarly incompatible elements (Niu and O'Hara, 2009). In primitive MORB prior to plagioclase on the liquidus both Eu/Eu* >1 and Sr/Sr* > 1, pointing to the Earth's MORB mantle

must have an inherited positive anomaly (i.e., excess Eu vs. Sm and Gd; and excess Sr relative to Pr and Nd. The significant linear correlation of Eu/Eu* with Sr/Sr* defined by the lunar samples (including glass beads from Saal et al., 2008), pointing to the significant plagioclase effect in the lunar petrogenesis. However, their trend differs from the MORB crystallization, and is more consistent with a mixing trend between incompatible trace element enriched samples with very low Eu/Eu* < 1, and ~ 0.2) and almost pure anorthositic rocks depleted in all the incompatible elements. As the lunar Eu/Eu* and Sr/Sr* do not correlate with MgO as in MORB, and in fact do not correlate with any major elements nor trace elements abundances except the inverse relationship with incompatible elements (see Figs. 4 and 5). The mixing can be (1) the KREEP-like melt mixes with the anorthositic cumulate in varying proportions or (2) mechanical things between the two by the near surface processes.

Figure 8.

Al_2O_3 (wt.%) variation diagrams of major element oxides (TiO_2, FeO, MgO, CaO in wt.%), minor element oxides (Na_2O, K_2O, P_2O_5, Cr_2O_3 in wt.%), representative trace elements (Ni, Co, Ce, Sr and Eu in ppm) and Sr/Sr* and Eu/Eu* ratios of representative lunar surface samples (regoliths) on Apollo and Luna sites (Lucey et al., 2006). The grey bands highlight where the KREEP-like material present with enriched incompatible elements and stronger negative Sr and Eu anomalies.

Figure 9.

Frequency of concentrations of Ti (as TiO_2) determined by gamma ray spectrometry across the lunar surface (data from Prettyman et al., 2006). This figure is based on the 2-degree bin data set where each of the 11306 results represents an average TiO_2 content in ~2 billion tonnes of regolith.

Figure 10.

Frequency of concentrations of Fe (as FeO) determined by gamma ray spectrometry across the lunar surface (data from Prettyman et al., 2006). This figure is based on the 0.5-degree bin data set where each of the 259200 results represents an average TiO_2 content in ~100 million tons of regolith.

Figure 11.

Al_2O_3 wt.% and FeO wt.% in Apollo 15 samples based on sources listed in Appendix 2. Salient features are the strong negative correlations between Al_2O_3 and FeO within both the highland and mare-derived samples reflecting the fact that the principal mineralogical variation is a balance between the relative amounts of the dominant minerals, plagioclase and the sum of olivine and pyroxene. Within the highland sample group only those samples with Al_2O_3 greater than ~24% are likely to display positive Eu anomalies (O'Hara, 2000, Fig 2 inset); compositions close to the average, or close to the bulk impact melts, have negative Eu anomalies. The average of the highland samples is shown by a cross, H, which is close to the field of compositions of bulk impact melt samples which can be expected to represent excellent mechanically mixed averages of large volumes of lunar highland crust in the Imbrium target material. H is also close to the field of soil compositions developed by the subsequent averaging process of regolith formation on the Apennine Front (average F). These soil compositions may contain a small admixture of mare components added by lateral scattering after mare emplacement.

Figure 12.

Concentrations of Fe (as FeO) and Al (as Al_2O_3) determined by gamma ray spectrometry across the lunar surface (data from Prettyman et al., 2006). This figure is based on the 5-degree bin data sets where each of the 1790 results represents average FeO and Al_2O_3 content in ~10 billion tons of regolith.

Figure 13.

This figure is based on 1790 averages of ~1010 (10 billion) metric tons of regolith in each observation of the concentration of CaO and Al_2O_3 (Prettyman et al., 2006) compared with data from the Apollo 11 and 15 mare basalts. 68415 has no Eu anomaly and plots at 28.5%Al_2O_3, 16.3% CaO towards the upper edge of the main concentration of results. The paucity of compositions close to anorthite where positive Eu anomalies are guaranteed, is evident. Equally conspicuous is the paucity of compositions overlapping the field of alleged primary magmas in the Maria. Average compositions of mafic glasses generated and scattered by impact to the Apollo 16 site (Zeigler et al., 2006) include a high-Al_2O_3 basalt group (HABG, blue diamond), probably derived from Mare Nectaris 220 km to the east, which plots among basaltic compositions expected to crystallize plagioclase before pyroxene. A low-Ti basalt group (LTBG, yellow diamond), probably also derived from Mare Nectaris, plots with the more feldspathic hand specimens and more mafic soils from the Apollo 15 site. A high-Ti basalt group (HTBG, orange diamond), identified as representative of a widely exposed mare surface in Mare Tranquillitatis 300 km to the north, has 8.6% TiO_2, 17.8% FeO and plots very close to experimentally generated plagioclase-saturated cotectic liquids from Apollo 11 (and 17) samples. Scattering for these distances implies substantial impacts and averaging of a significant depth of mare material. Instrumental uncertainties may underlie in part the spread of results to the peraluminous side of the line connecting ‘anorthite’ to the origin, which otherwise reflects a contribution from alkali feldspars.

Figure 14.

The frequency of Al/Si ratios in the lunar surface (data from Lawrence et al., 1998, Prettyman et al., 2006) peaks close to Al/Si ~ 0.65, with a low frequency of values above 0.8 at which a positive Eu anomaly can be expected. Values of Al/Si < 0.27 matching the putative primary magma compositions are scarce. This histogram is based on 1790 averages of the top ~ 0.3m of regolith in areas of ~22500 km^2, or an average of ~1010 (10 billion) metric tons of regolith in each observation.

Figure 15.

SiO_2 vs. Al_2O_3 plot showing lunar mare-derived meteorites (stars) compared with fields of data (shaded) for some Ti basaltic materials from the Apollo 15 site (from left to right, hand specimens, experimental plagioclase-saturated cotectic liquids, mare surface regoliths and breccias). Filled diamonds show average compositions of mafic glasses generated and scattered by impact to the Apollo 16 site (Zeigler et al., 2006) and include a high-Al_2O_3 basalt group (HABG), which plots among basaltic compositions expected to crystallize plagioclase before pyroxene. A low-Ti basalt group (LTBG), plots with the more feldspathic hand specimens and more mafic soils from the Apollo 15 site, among the cotectic liquids. A high-Ti basalt group (HTBG), plots very close to experimentally generated plagioclase-saturated cotectic liquids from Apollo 11 and 17 samples (not shown). Broken lines at fixed Al/Si ratio facilitate comparison with O’Hara (2000 Fig 2) and with Figure 13 above. Apollo 15 sauces from Appendix 2.

Figure 16.

Frequency of concentrations of Al (as Al_2O_3) determined by gamma ray spectrometry across the lunar surface (data from Prettyman et al., 2006). This figure is based on the 5-degree bin data set where each of the 1790 results represents an average alumina content in ~10 billion tons of regolith. The assumption that the mare basalts (black) were primary magmas generated the LMO hypothesis and a requirement for a plagioclase cumulate crust (pale grey) with positive Eu anomalies. The petrogenesis of the bulk of the exposed material (dark grey) may have been under-addressed.

Figure 17.

The key feature is the wide variability in $Mg^{\#}$ from 80 to 40 at high Al_2O_3 (A), low MgO (B) and low FeO (C). One possible explanation might be control of liquid compositions by evolution along a plagioclase-saturated cotectic under yet-to-be-determined conditions of P, T and volatile contents which placed that cotectic in highly feldspathic bulk compositions.

Appendix 1: Data sources

Data for Figures 2, 3, 4, 5, 7 are exclusively from Lucey et al. (2006). Data for Figure 5 are from Lucey et al. (2006) for data points and derived from Heiken et al. (edited) (1991). Data for Figure 7 are from Lucey et al (2006), Saal et al. (2008), Niu and O'Hara (2009).

All other chemical data for rocks, clasts, breccias and soils have been drawn from Cuttita et al. (1973), Drake and Klein (1973), Dowty et al. (1973), Duncan et al. (1975), Dymek et al. (1974), Laul and Schmitt (1973), Lindstrom et al. (1977), LSPET (1972), Mason et al. (1972), Maxwell et al. (1972), Nava (1974), Powell et al. (1973), Rhodes and Hubbard (1973), Roedder and Weiblen (1972), Rose et al. (1975), Ryder and Schuraytz (2001), Ryder and Steele (1988), Vetter et al. (1988), Wänke et al. (1972, 1973, 1977) and Willis et al. (1972). Powell et al. (1973) analysed fused beads prepared from small lithic fragments. The fusion technique is not specified but there is a strong suggestion of substantial soda loss by volatilisation during preparation of these beads in view of the very low soda contents reported. Roedder and Weiblen (1972) reported silica-poor inclusions in the olivines of 15555, 34 which may represent samples of trapped early liquids.

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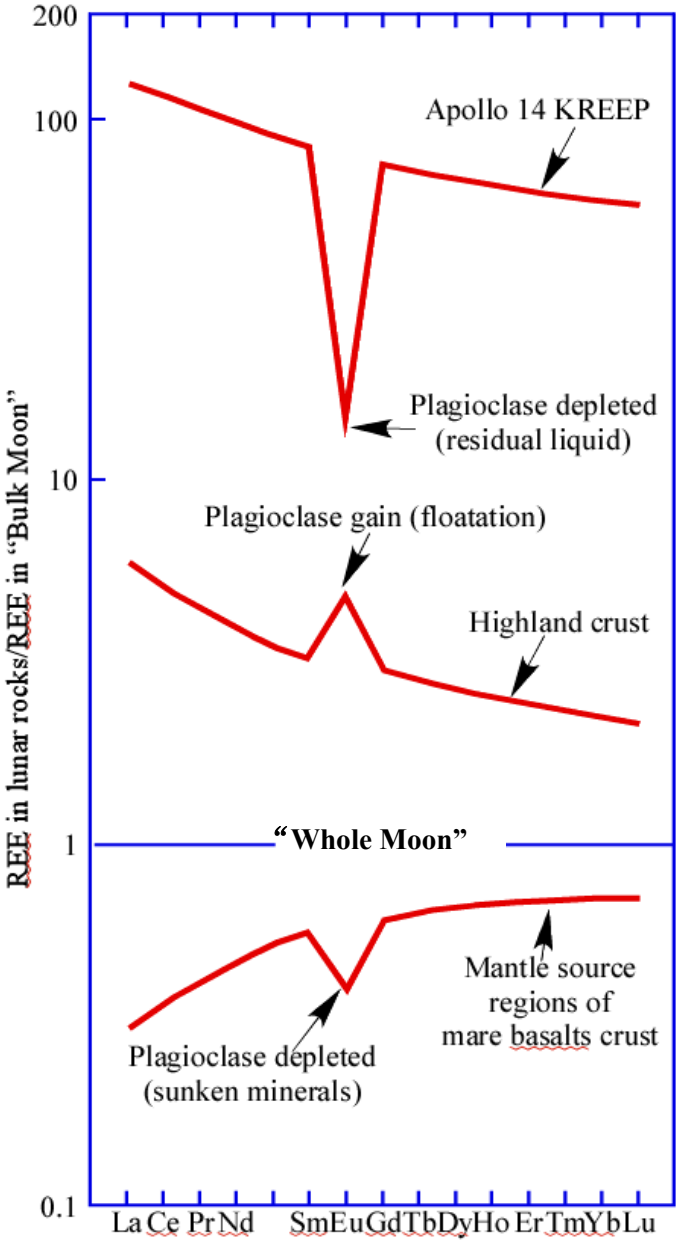
Figure 1. (O'Hara & Niu, 2015)



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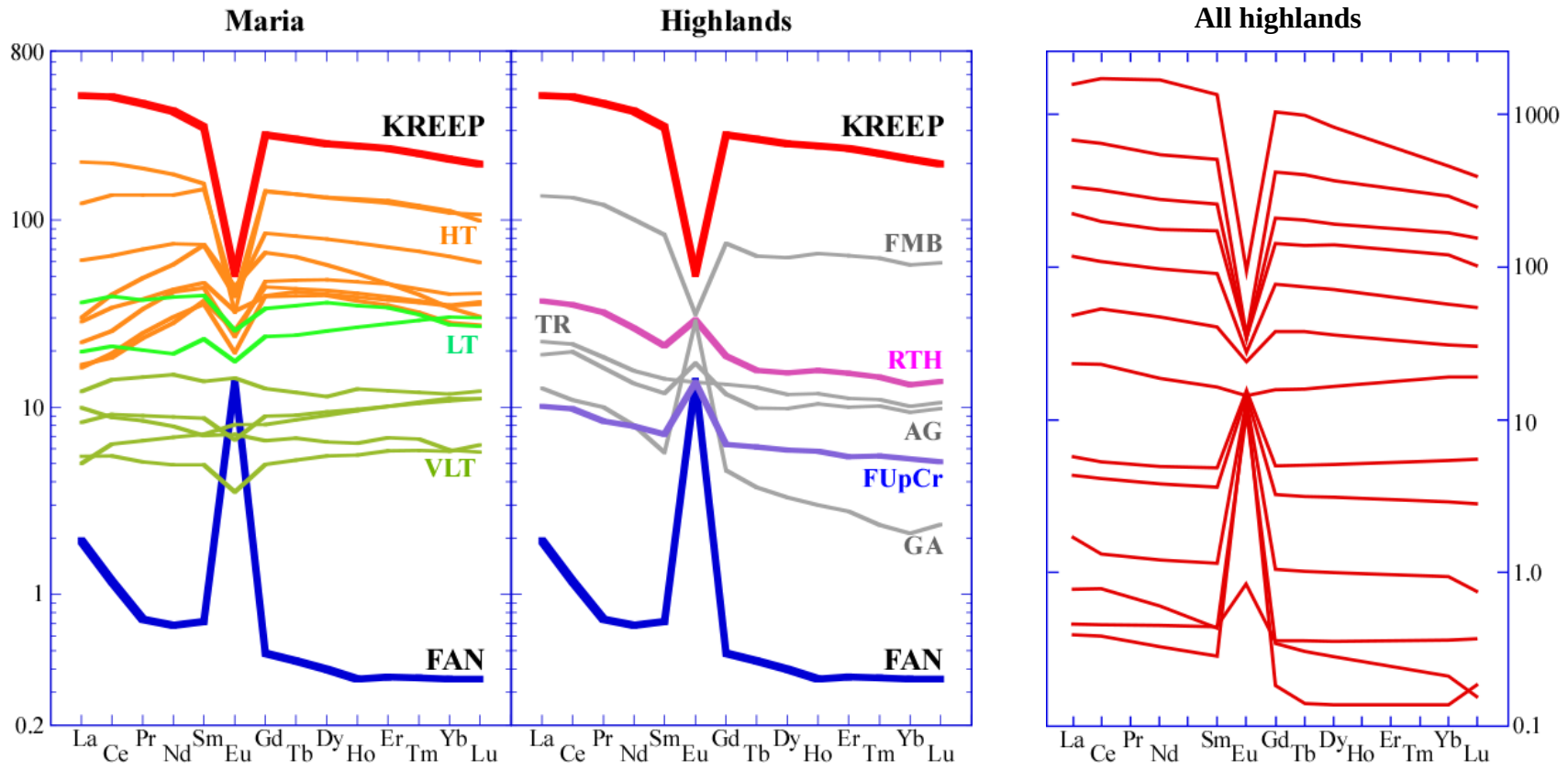

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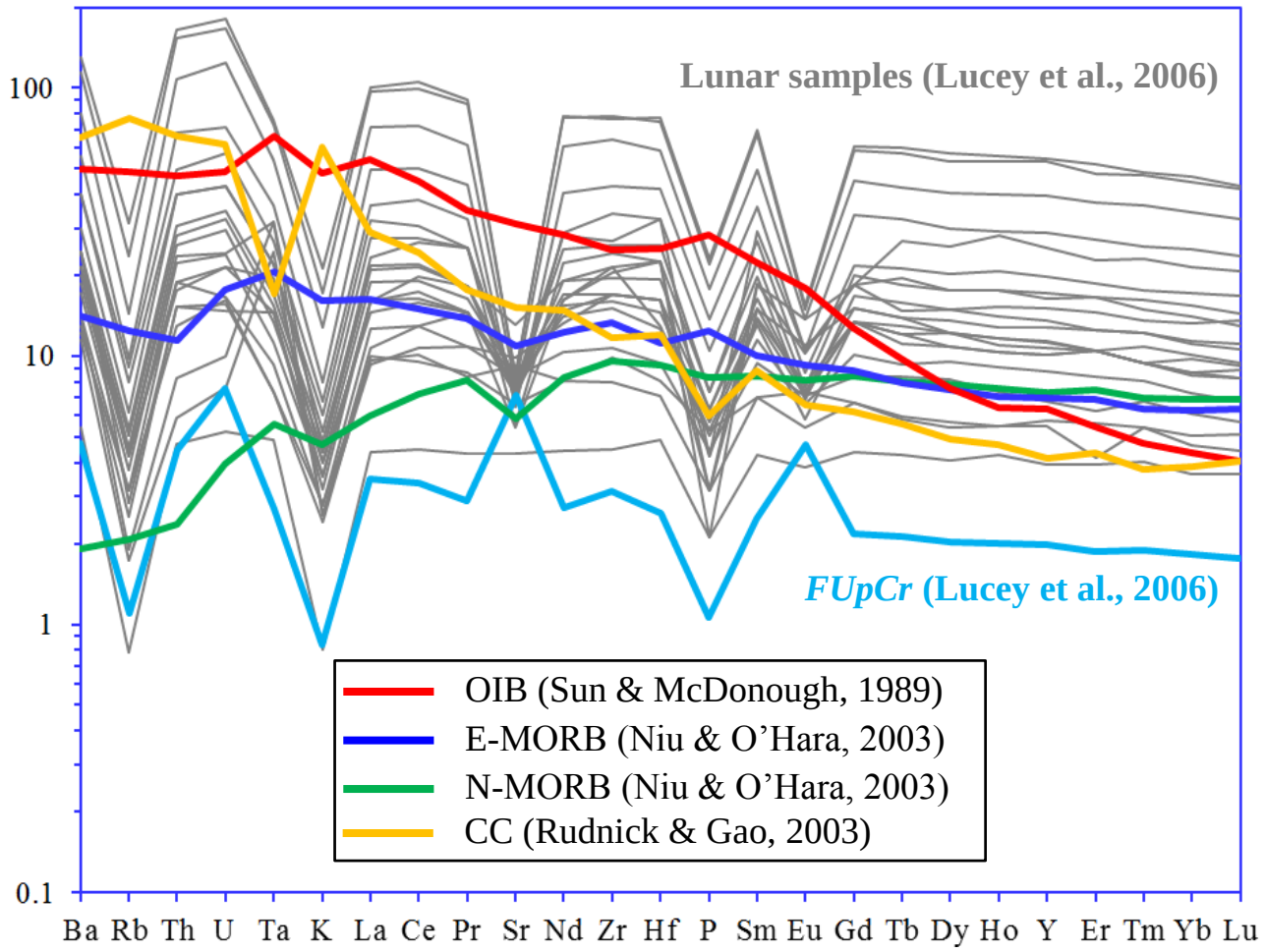

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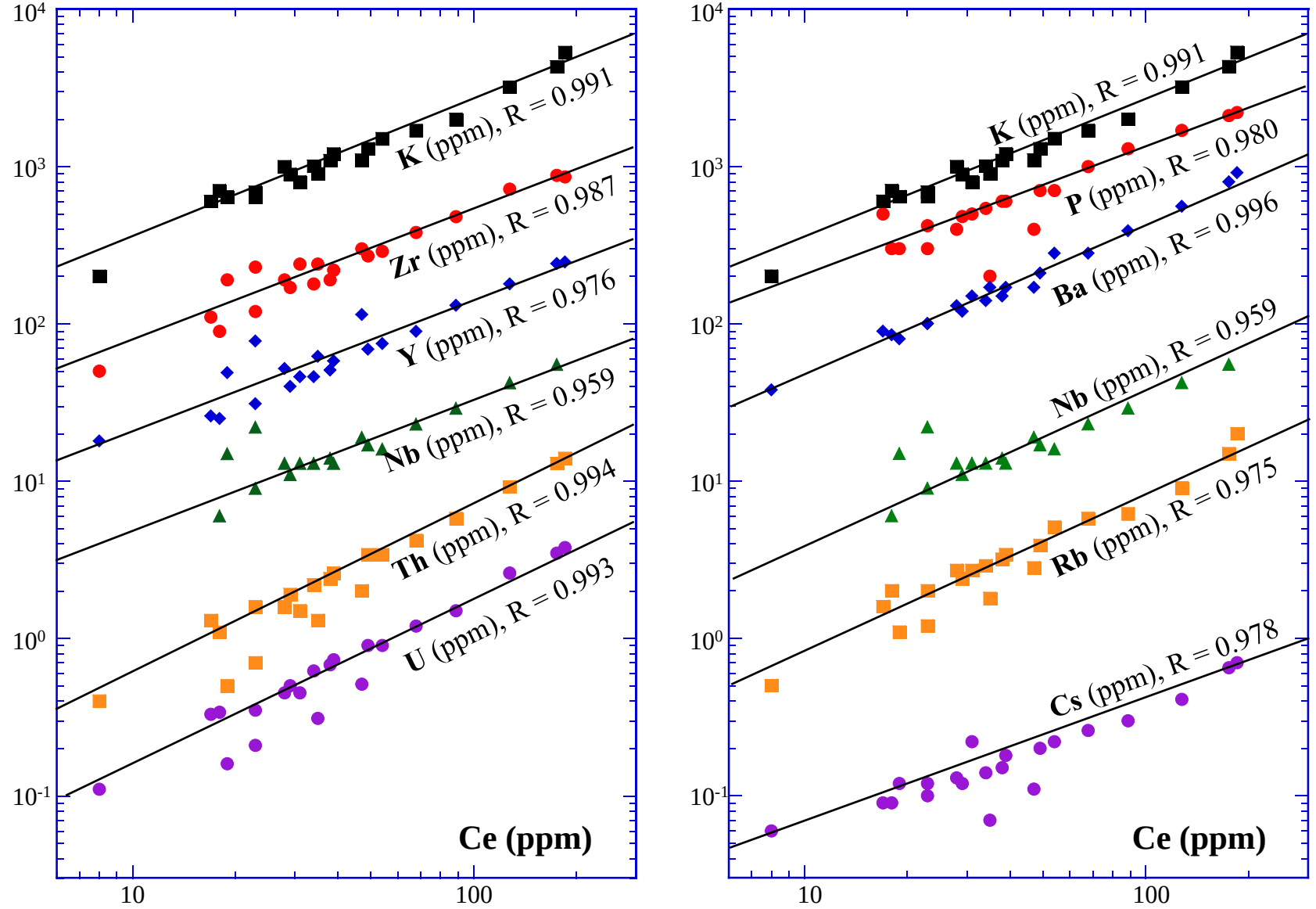

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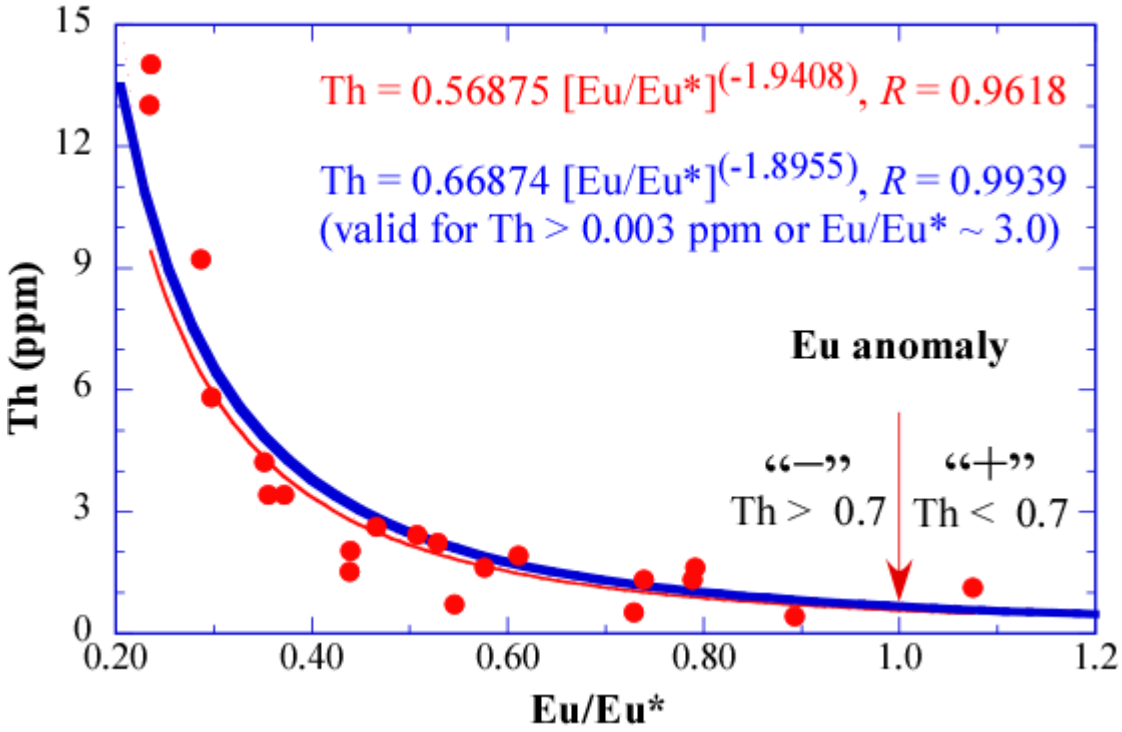

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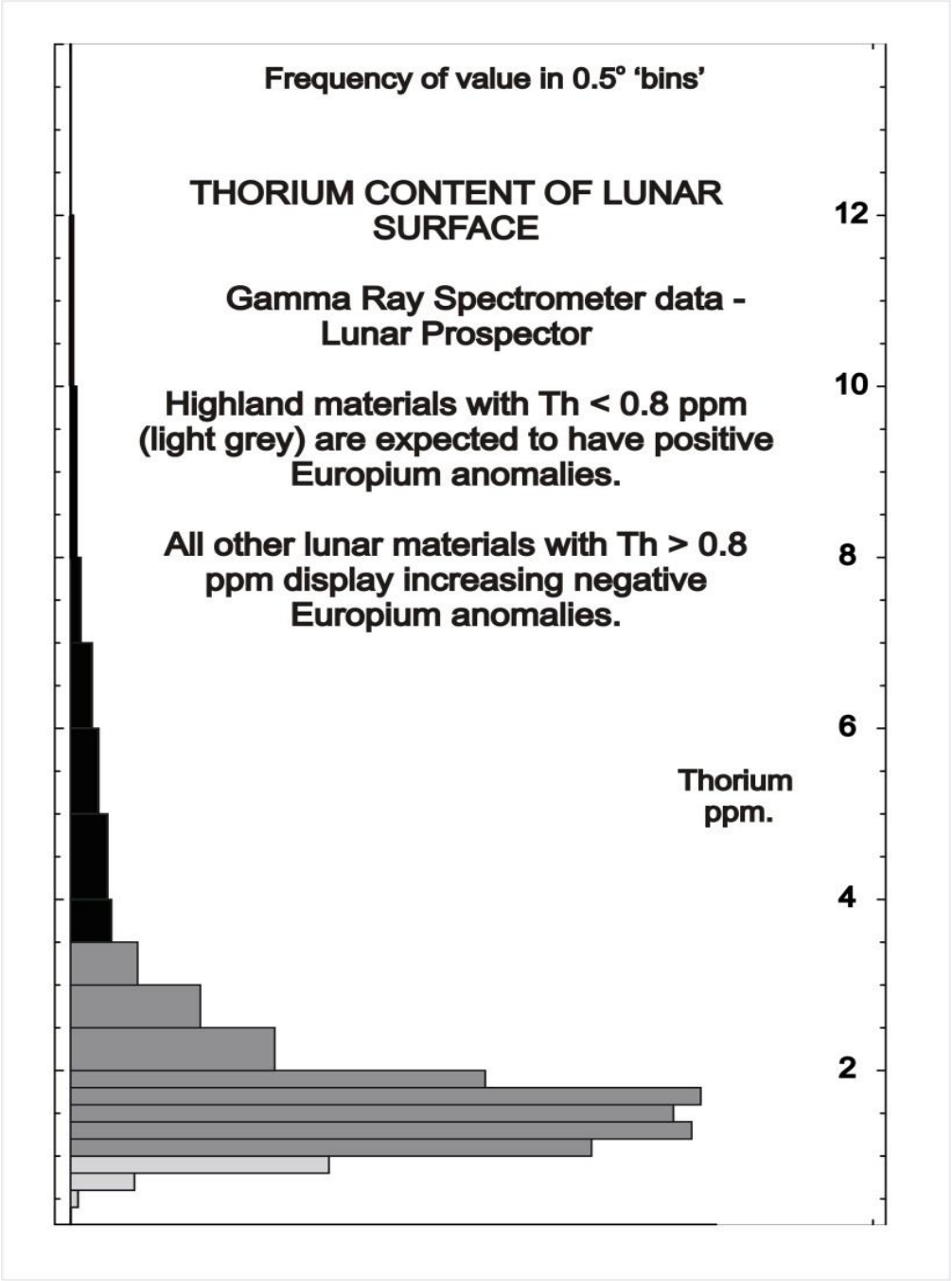

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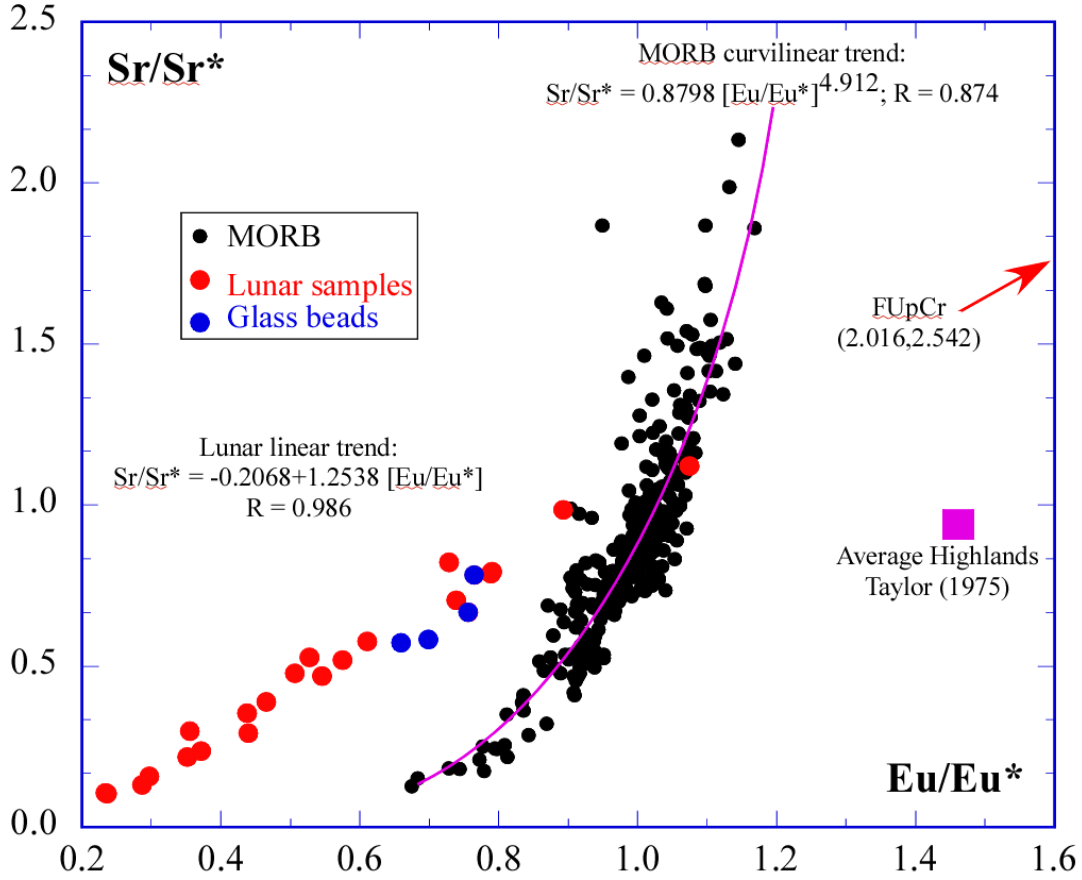

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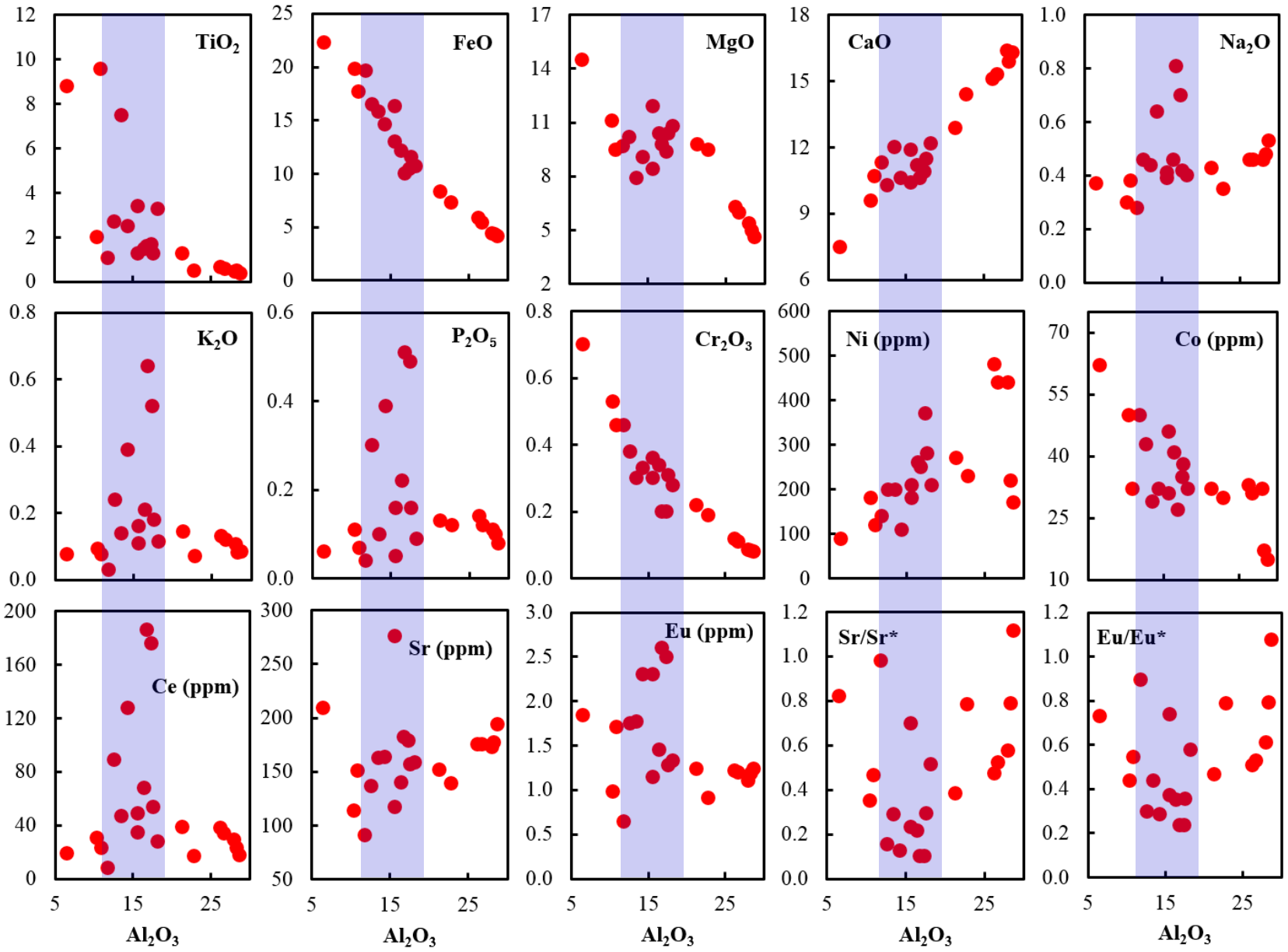

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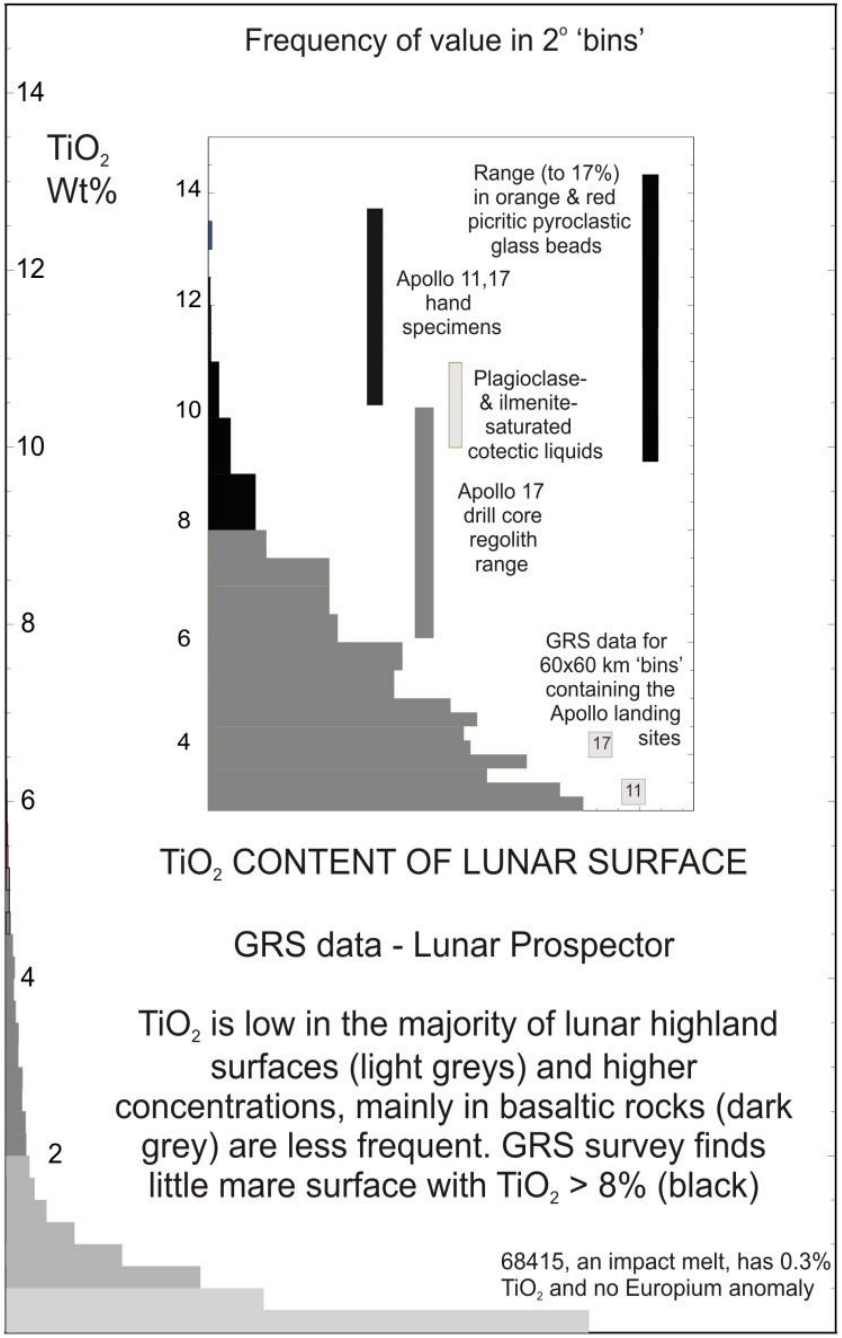

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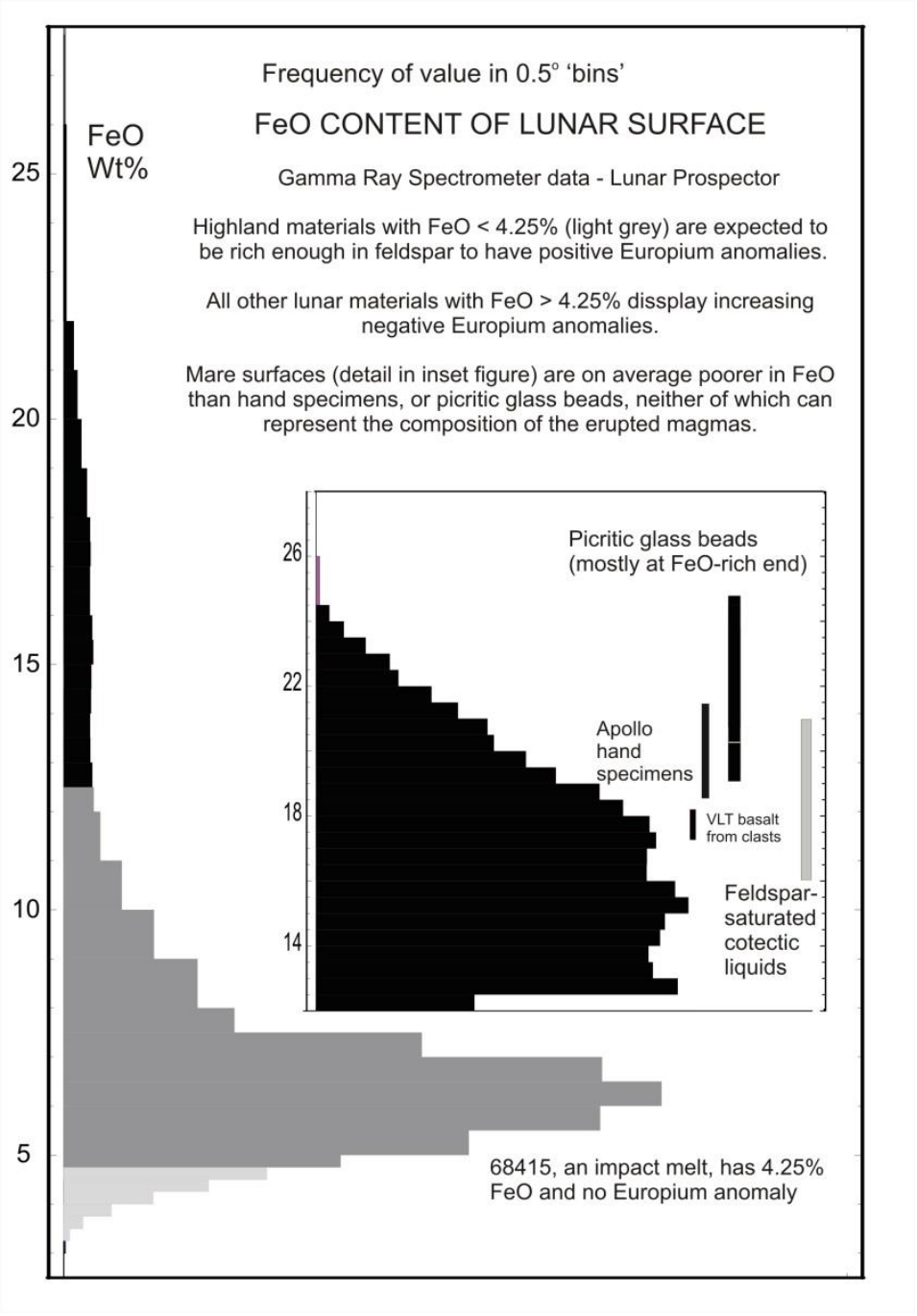

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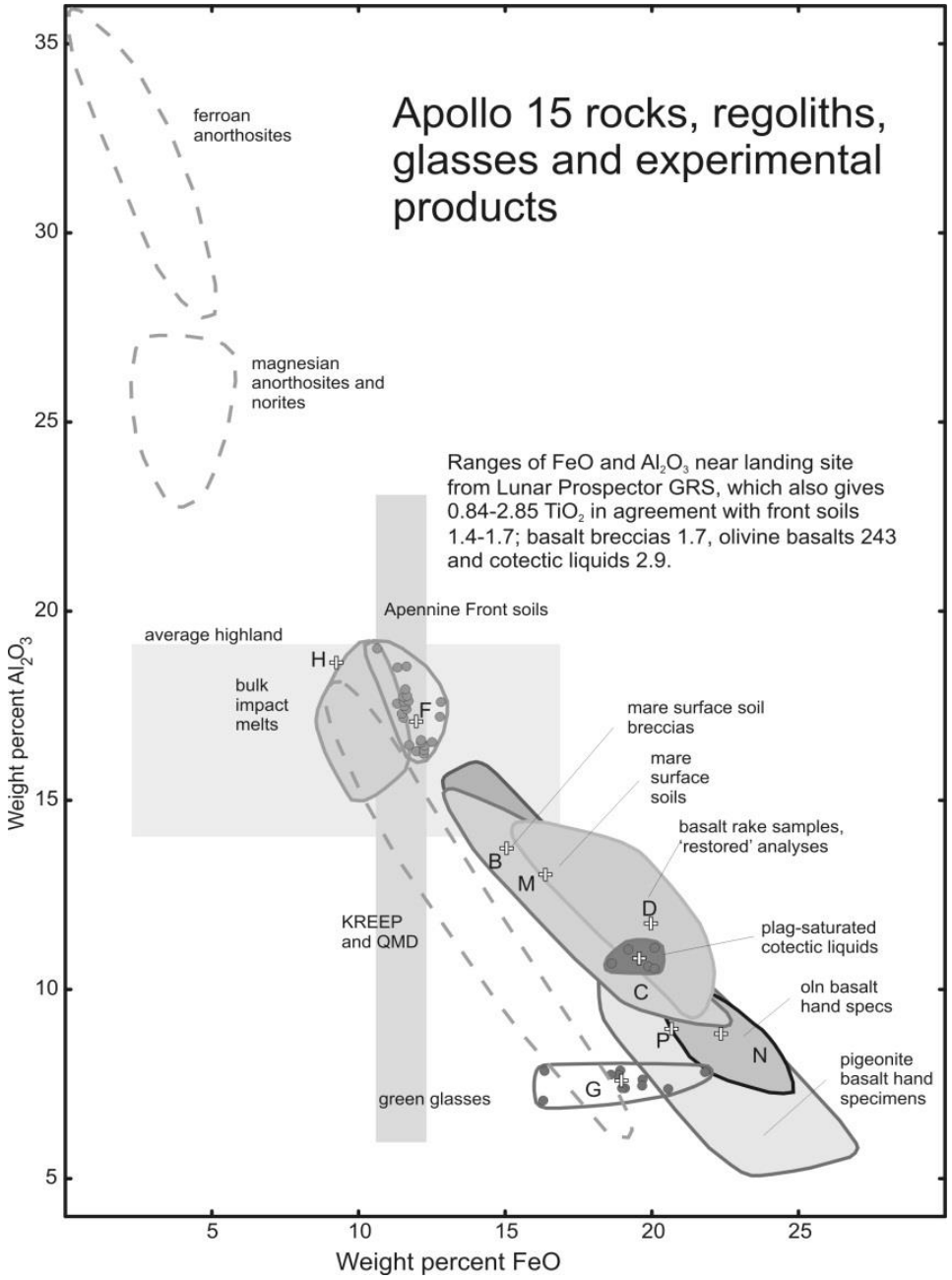

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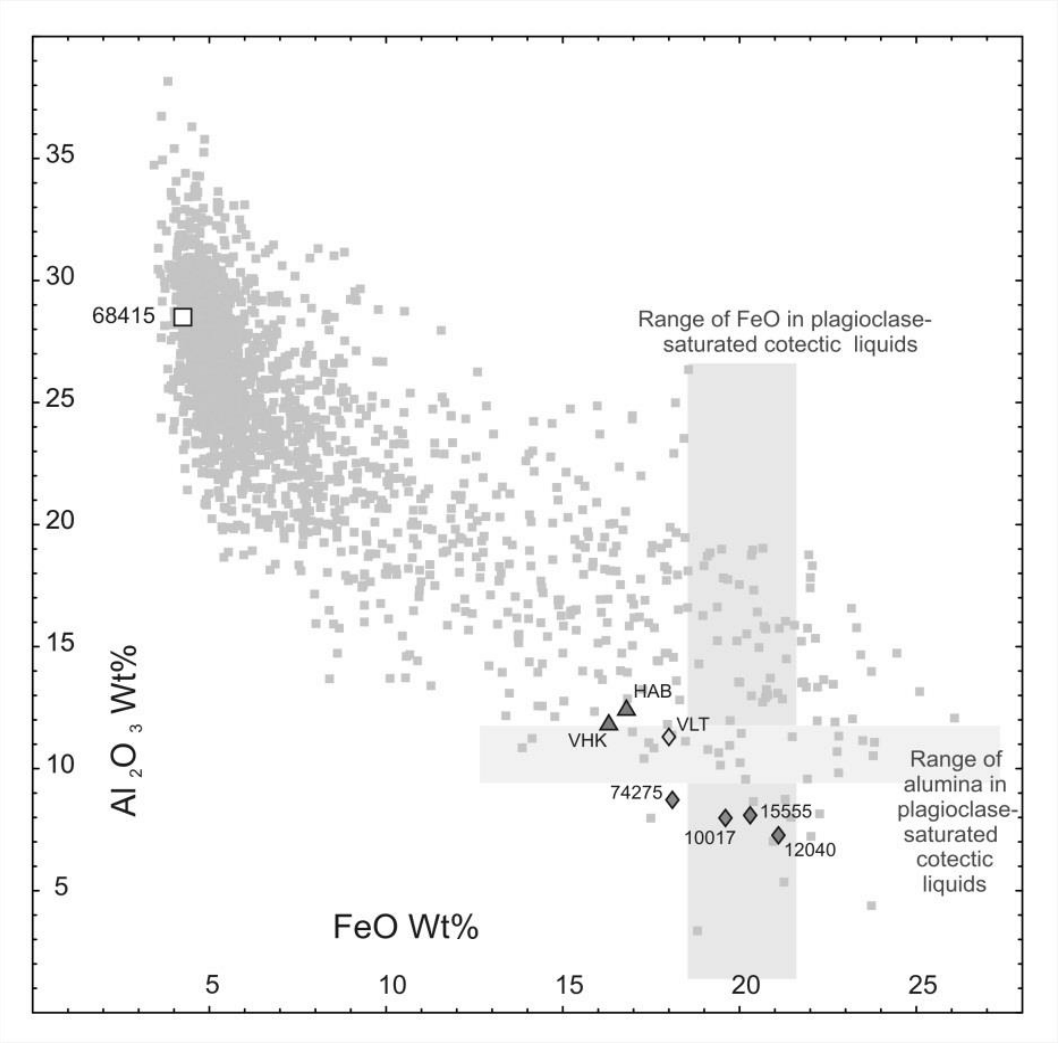

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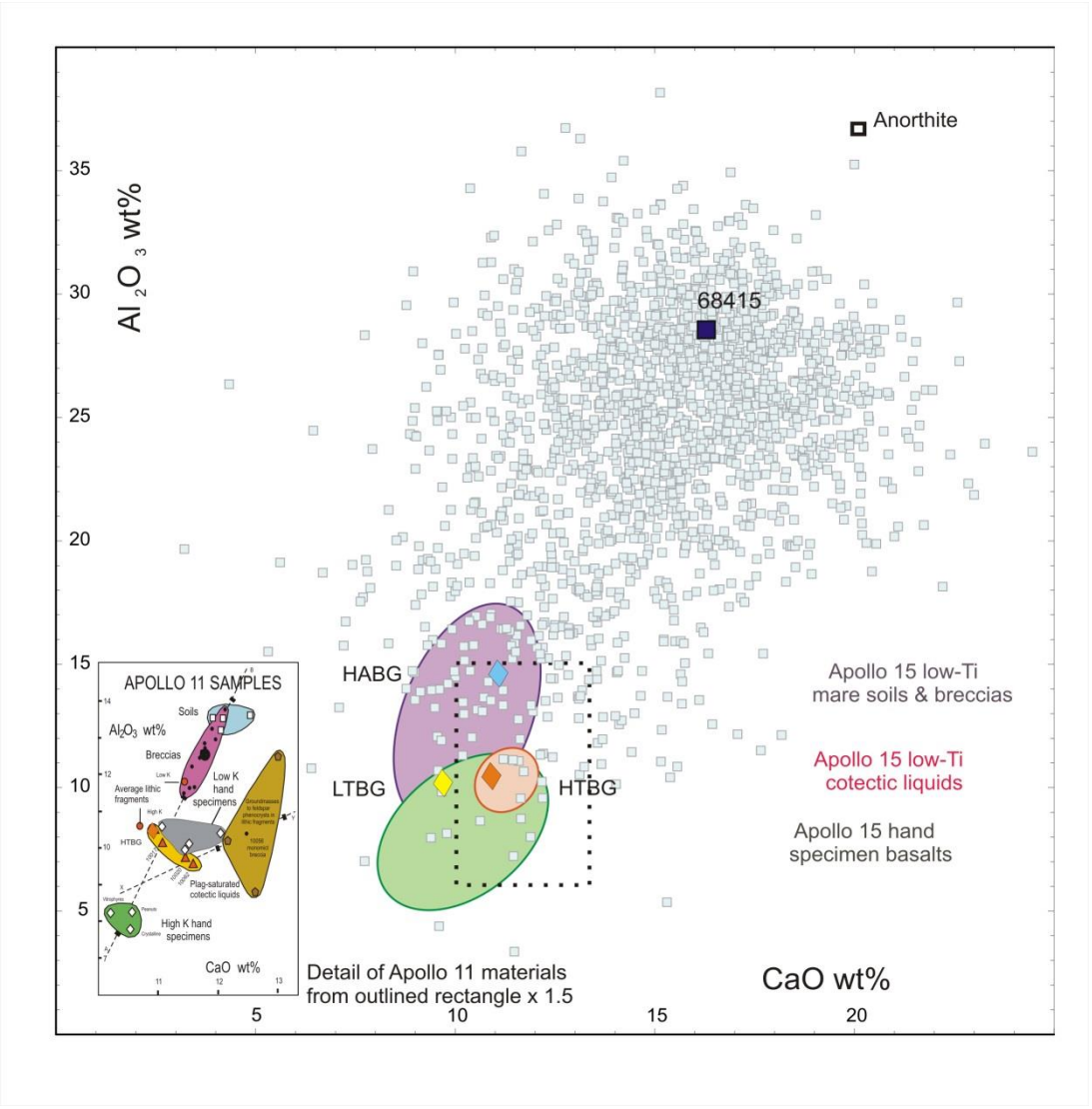

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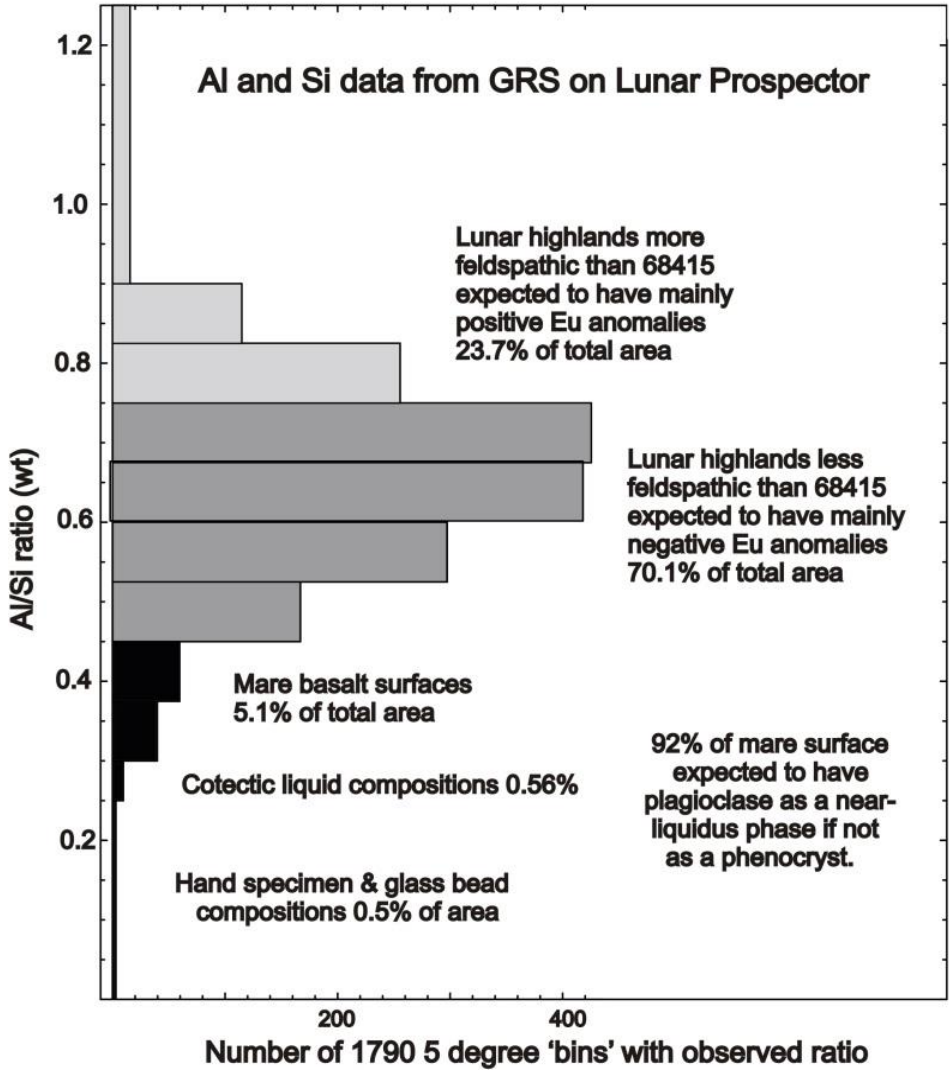

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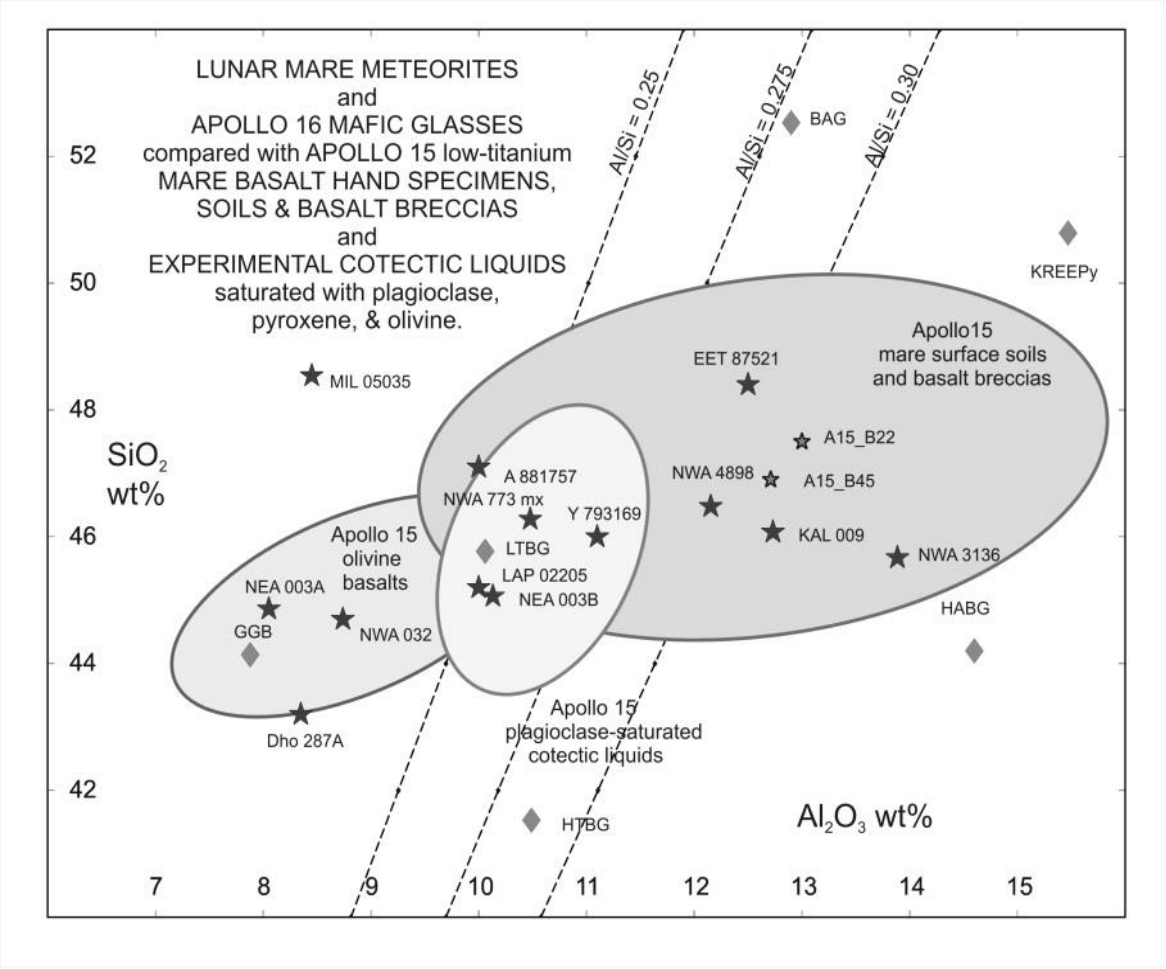

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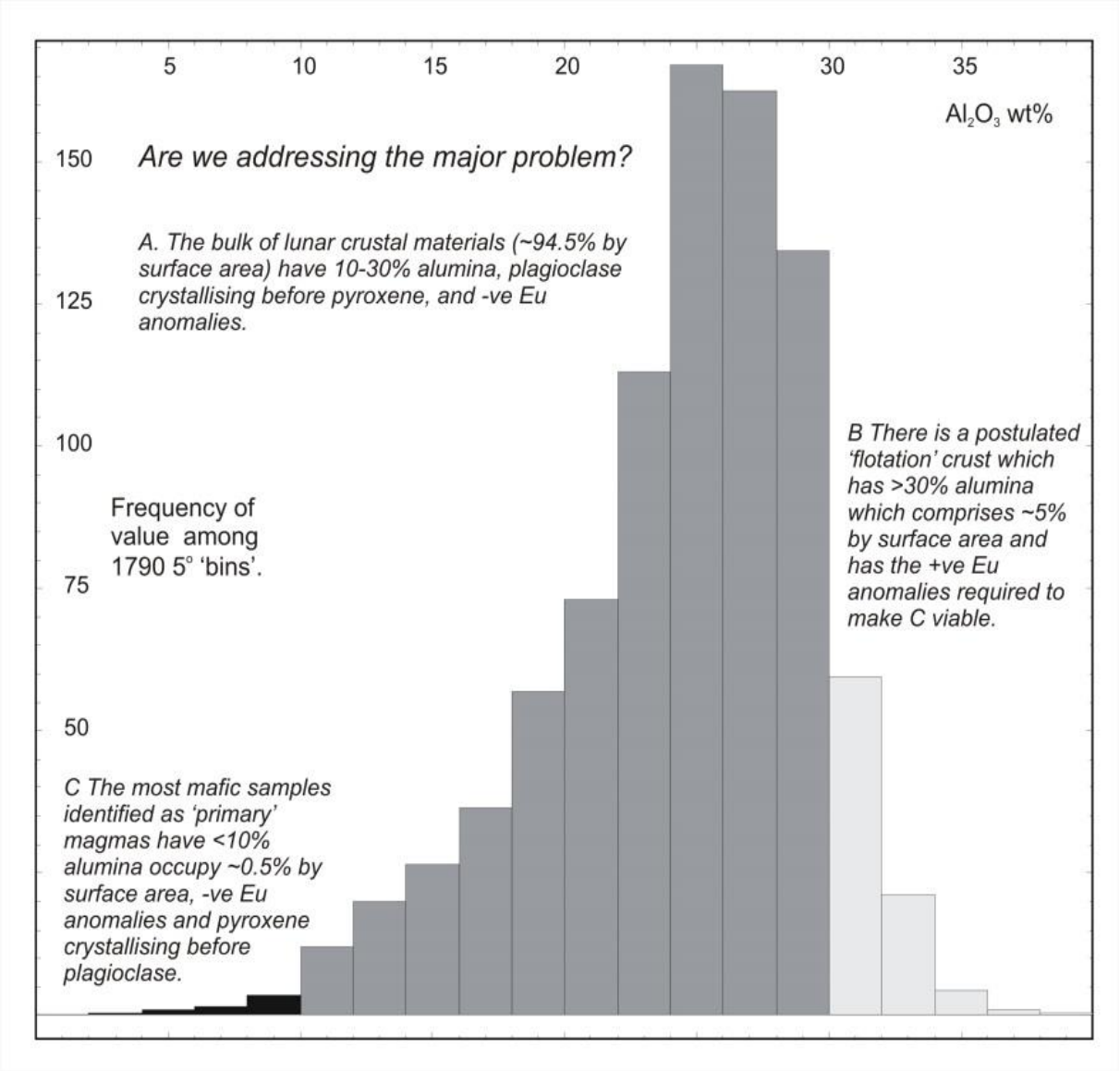

Figure 17. (O'Hara & Niu, 2015)